

SCIENCE

MARCH 17, 1950



THE SIZE OF SILICONE MOLECULES

THEODORE G. ROCHOW AND
EUGENE G. ROCHOW

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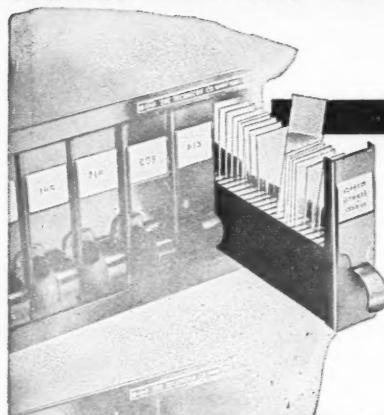


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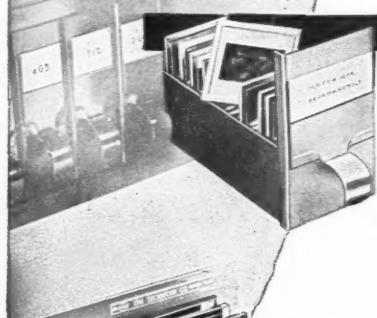
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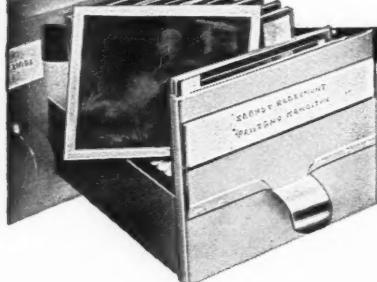
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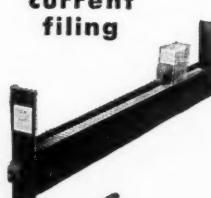


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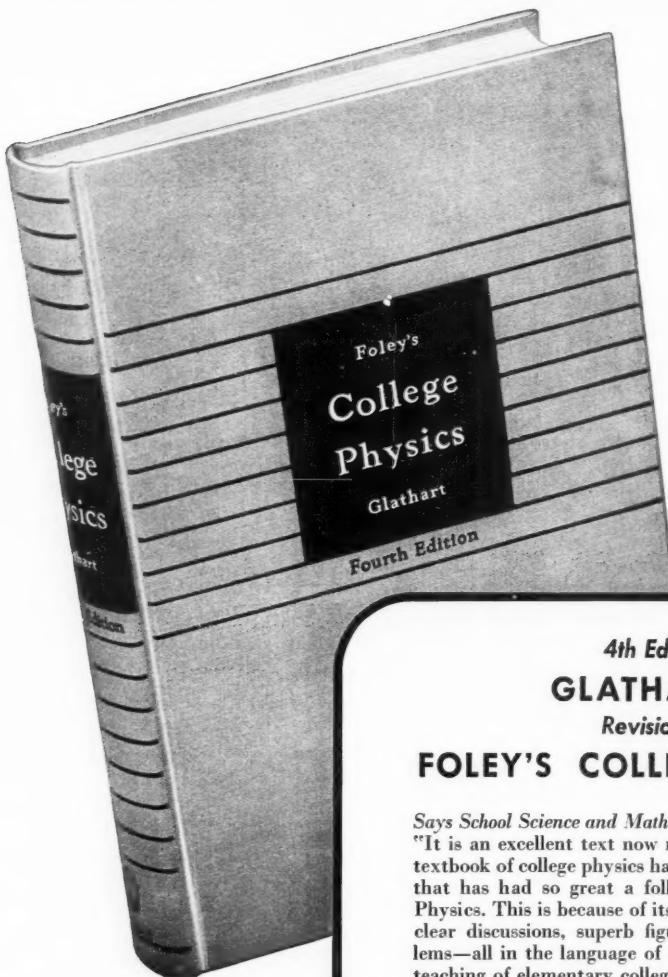
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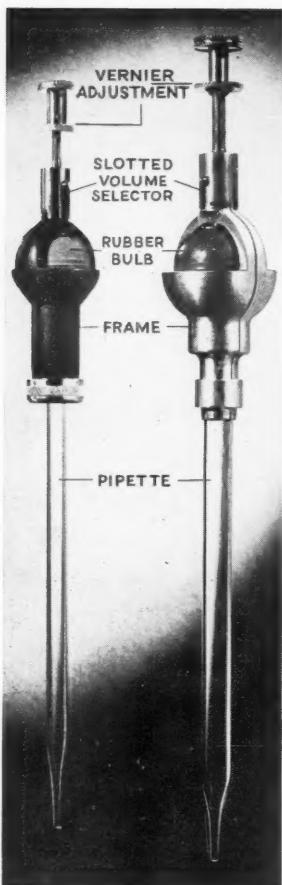
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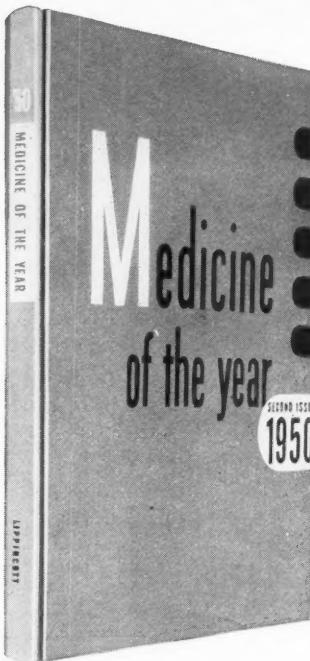
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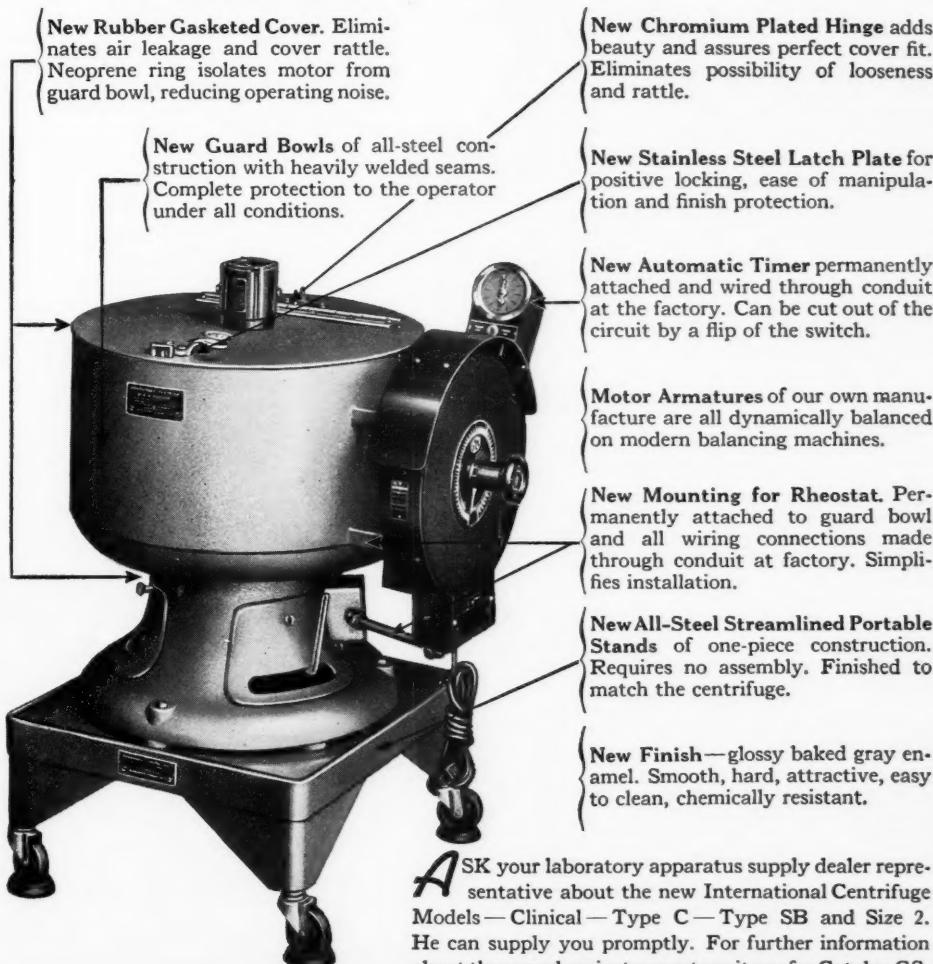
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The Size of Silicone Molecules

Theodore G. Rochow and Eugene G. Rochow

*American Cyanamid Company, Stamford, Connecticut, and
Department of Chemistry, Harvard University, Cambridge, Massachusetts*

THE SILICONE POLYMERS, which are synthetic organosilicon resins, oils, and elastomers based on frameworks of alternate silicon and oxygen atoms, are well enough known by now so that their composition and their modes of preparation are fairly well understood. Very little is known, however, about the size and configuration of the large molecules that make up such silicone polymers, beyond the fact that such macromolecules have remarkably little attraction for each other. The question that gives rise to the present discussion is whether or not the methods of resinography might be applied to the problem, and what information might so be gained.

Resinography is the graphic study of resins, involving the use of the microscope and all of its adjuncts in techniques somewhat similar to those of metallography and mineralography. As applied to the study of fabricated plastics, for example, such study yields information about the kind and shape of fillers, the distribution of strengthening materials, and the degree of wetting of such included bodies by the binding resin (5). The particular technique of resinography that is of most value in the present instance, however, is the electron microscopy of fracture surfaces of synthetic high polymers (6). By fracturing a cooled solid specimen and then preparing a replica of the surface, the commensurate irregularities and discontinuities of the fracture surface may be shown by micrographing the replica at high magnification in the electron microscope.

To show the features most plainly, a particular kind of replica must be made. The fractured surface first is coated with a 5 percent aqueous solution of gelatin, which is stripped off after drying. Silica is evaporated onto the gelatin replica in a vacuum evaporator, and the gelatin is dissolved away from the silica by water, to leave a positive "print" of the surface in silica. In order to emphasize some surface features in elevation, it is possible to "shadow" the replica by depositing uranium by evaporation at a small angle, giving the same effect as one gets in observing hilly terrain in the light of a setting or a rising sun. The final replica then is electron-micrographed at magnifications up to 20,000. Selected micrographs are enlarged photographically as required by the fineness of detail in the micrograph and the distance of viewing the final picture.

The technique of embrittling a high polymer and preparing a replica of its fracture surface is generally similar to one of the methods employed in 1944 by Ladd in his electron-microscopical studies of colloidal carbon in vulcanized rubber (2). He cooled a block of tire tread stock in a bath of acetone-dry ice and cracked the block in two. A negative replica of the broken surface was made of softened De Khotinsky cement or polystyrene. This was stripped off when hard and a drop of 2 percent Formvar solution in ethylene dichloride was placed on the negative impression and allowed to harden. The cement or polystyrene was dissolved away, leaving the Formvar positive replica to be electron-micrographed. Ladd obtained poor definition of the carbon particles and experienced difficulty in interpreting the micrographs. Apparently he abandoned the method in favor of pressing out a thin film and vulcanizing it in this condition. Such a method, of course, requires uncured material to begin with and assumes the toleration of probable distortion of the morphology.

Rochow and Rowe (6) were more fortunate in using a water-dispersible material like gelatin because it was readily peeled from the fracture surface and yet wetted the surface sufficiently to form a good impression. They subsequently applied this technique to vulcanized tire tread stock¹ after taking the further precaution to immerse it in liquid nitrogen to be certain of obtaining a brittle fracture. Fig. 1 is an electron micrograph of a gelatin-silica bireplica of the carbon-reinforced, cured tire tread stock. The carbon particles are shown singly and in aggregates. The particles are shown in three manifestations: (1) as black disks representing carbon particles that were pulled from the fracture surface and remained with the final replica, (2) as elevated gray disks representing carbon particles that remained at or very near the replicated fracture surface and (3) as depressed gray disks representing carbon particles that went with the mate of the fracture surface. The gray matrix represents the cured, natural rubber polymer.

One probably sees no structure in the matrix as shown at this magnification, and by halftone reproduction. If, however, the micrograph is enlarged to 100,000 \times , the matrix may be perceived to be particu-

¹ Material by courtesy of A. R. Davis, Research Laboratories, American Cyanamid Company, Stamford, Connecticut.

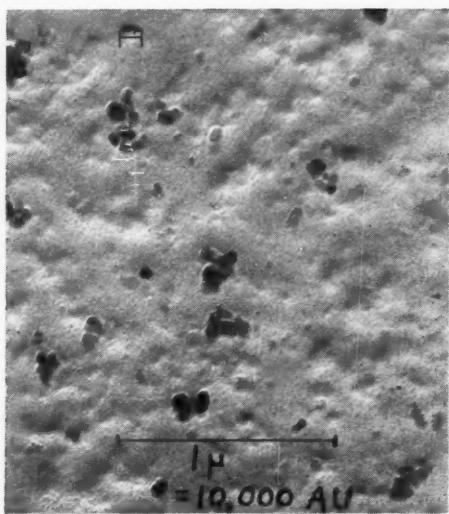


FIG. 1. Bireplica of fracture surface of black natural rubber. Positive print. Magnification 35,700 \times .

late, as shown in Fig. 2. The smallest particles shown in the picture are about 0.65 mm in diameter, those occurring most frequently being about 1.3 mm and

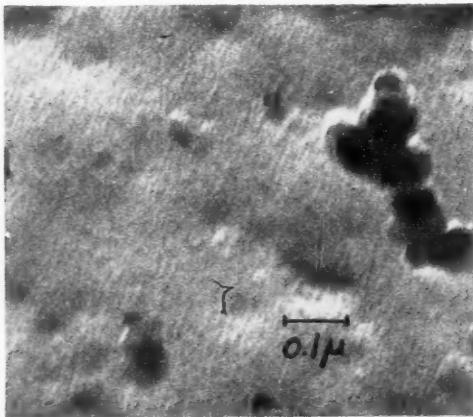


FIG. 2. "Close-up" of fracture surface of black natural rubber. Magnification 100,000 \times .

most of the largest about 2.0 mm. Since the magnification is 100,000 \times , these diameters, expressed in angstroms, are 65,130 and 200, respectively.

It is a natural assumption that the discontinuities which appear on a fracture surface reflect weaknesses in the cohesion of the structural units of the solid, i.e., the substance fractures between the structural entities. In a single polymeric phase, such as the rubber elastomer shown in Fig. 2, the structural entities are

assumed to be the macromolecules. Using the values just given for the diameters of typical particles, and the first of the methods of calculation given later in this discussion, the macromolecular weights of cured natural rubber are found to vary from 81,000 to 2,200,000, with most of them in the region of 650,000. Values determined on uncured ("soluble") natural rubber by older methods, such as measurement of viscosity or osmotic pressure of solutions, vary between 68,000 and 435,000 (1) and less and more. It appears, then, that the discrete particles are the macromolecules themselves. It is assumed that the rubber phase breaks by tearing apart such molecules rather than by splitting the molecules themselves. Expressed otherwise, the intermolecular (Van der Waals') forces are weaker than the chemical bonds which hold the macromolecule together, as would be expected and as indeed is inherent in the fundamental concept of molecules.

Application of this resinographic method of molecular size determination is of particular interest in silicones because of their peculiar properties. Silicones always are polymeric (as is silica itself), but the size of the molecules and the particular mechanism of their growth have been in doubt. It had been assumed in the early work that the processes leading to increase of molecular weight were interrupted by various agencies not always related to the purity of the intermediates, and this interruption was sup-

TABLE I
MOLECULAR WEIGHT OF DIMETHYLSILOXANE ELASTOMER*

Fraction	Percent of polymer	Avg mol wt
A	17	2,800,000
B	14	1,500,000
C	18	610,000
D	27	290,000
E	15	much less than 290,000

* Taken from Scott (9).

posed to leave molecules of decidedly low molecular weight, at least by the standards of the chemistry of organic high polymers. Such a view gave an easy explanation for the low melting point of diphenyl silicone, the comparatively low tensile strength of silicone rubber as ordinarily made, and the low change of viscosity with temperature in silicone oils (4). These are manifestations of the easy motion of molecules relative to one another, and, of course, a small molecular size contributes greatly to such ease of motion. However, the low boiling point of dimethyl polysiloxanes of known molecular weight and configuration (and indeed the low boiling points of most organosilicon substances) should have hinted that such relative ease of molecular motion and detachment was not due to low molecular weight but rather to inher-

ently low intermolecular attraction. Finally, when Scott (9) determined the molecular weights of five fractions of a representative silicone elastomer (Table 1), it was realized that the molecules of silicone rubber were indisputably large by ordinary standards—in fact, much larger than those reported for many common organic elastomers. The work of Wilcock (11) showed in turn that the flow properties of methyl silicone oils could be explained entirely in terms of low intermolecular forces, resulting in high molar volumes and a small energy requirement for motion of the characteristic unit of flow. It remained for Roth (7) to explain such low intermolecular attractions in terms of the exceedingly free motion of dimethylsiloxane units about the Si-O bond, such freedom being related in turn to the ionic character of such a bond.

It must be conceded, then, that the properties of silicone polymers are due to the unique combination of large molecules containing many points of relatively free motion, and that such properties therefore derive from the presence of siloxane bonds in the molecule rather than from the mere presence of silicon atoms. This has been demonstrated in very definite fashion by Sommer (10), who has shown a decided difference between polymers based on the Si-O-Si networks and those based on Si-CH₂-Si networks, despite the almost equivalent molecular weight.

When it was realized that the properties of silicone polymers were not due to limited molecular weight, and that some silicones might contain molecules of exceedingly large size, the actual size became of much interest. The only investigation of molecular weight until very recently was that of Scott, noted above. Scott dissolved a sample of dimethyl silicone elastomer in ethyl acetate and precipitated fractions of decreasing molecular weight by adding acetone progressively to the solution; he then determined the molecular weight by measuring the osmotic pressure of dilute solutions of the fractions by a dynamic method. Such a method naturally is limited to soluble polymers, and some question arises even then about whether molecules are broken apart by the process of solvation. The method is closed to many silicone resins which are no longer soluble in organic solvents after they are fully cured, but it also is desirable to know the size of the molecules in such resins. Nevertheless, Scott's measurements provide an opportunity to compare the results of the resinographic investigation with those of the more conventional dynamic osmometer method.

The first sample of silicone polymer chosen for test was a molded sheet of white silicone "rubber,"² prepared from dimethyl silicone elastomer and titan-

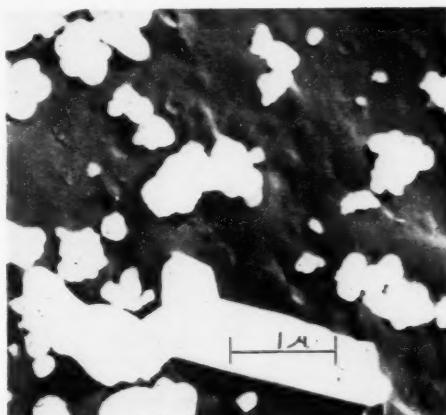


FIG. 3. Bireplica of fracture surface of white silicone rubber. Negative print of electron micrograph of polyvinyl + silica bireplica. Magnification 17,000 \times .

ium dioxide filler. A piece was cooled in liquid nitrogen and fractured. A silica replica was prepared by the method previously described, except that an 8 percent solution of polyvinyl alcohol was used instead of gelatin solution. Fig. 3 shows a micrograph of the replica. The white specks are particles of crystalline titanium dioxide filler, which also is represented by

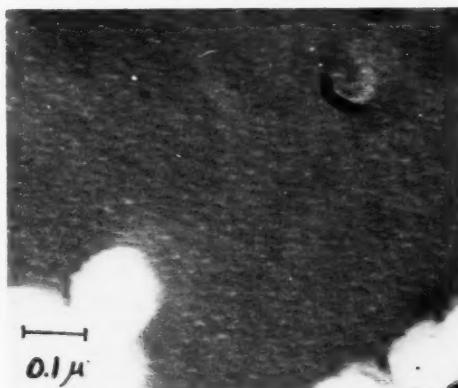


FIG. 4. "Close-up" of fracture surface of white silicone rubber. Magnification 100,000 \times .

the elevations and depressions caused respectively by particles on or near the fracture surface and those remaining with the fracture-mate. Thus we have a method for portraying the actual distribution of particulate substances from pigment-sizes down to and including reinforcing agents such as carbon black and dehydrated silica gel.

The gray background in Fig. 3 is the silicone matrix. At this magnification the matrix presents a grainy ap-

² Submitted by courtesy of the General Electric Company.

pearance, but no definite structure can be ascertained. At a magnification of $100,000 \times$ (Fig. 4) the graininess is more distinct, and is seen to be due to a particulate structure. In the electron micrograph, one millimeter corresponds to 100 Å in the original specimen, so that a direct measurement of the particle size becomes possible. The largest grains appear to be elliptical, and are about 0.93 mm by 3.7 mm. The smaller ones are more spherical, and are about 0.93 mm to 1.9 mm in diameter. Some still smaller grains may be seen, but measurement of them is difficult. For calculation, we may take a sphere of 1.0-mm diameter to be representative of many of the larger particles (but not the largest). One millimeter, as we have said, corresponds to 100 Å in the original, and the volume of a sphere 100 Å in diameter is $\frac{4}{3}\pi(50)^3$, or 524,000 Å³. We are justified only in calling it about 520,000 Å³.

We need now to translate this size into its more common terms of molecular weight, and one way of doing this is to determine the weight of a single macromolecule and multiply the weight (stated as the fraction of a gram) by Avogadro's number representing the number of molecules contained in one gram-molecular weight of a substance (6.02×10^{23} molecules). The specific gravity at the room temperature for the hard gum from which the white silicone rubber was compounded was measured to be 0.972. Thus the weight of one molecule of the particles with radius 50 Å (5×10^{-7} cm) is: $520,000 \times 10^{-24}$ cm³/molecule $\times 0.972$ g/cm³ $\times 6.02 \times 10^{23}$ molecules/mol = 304,000 g/mol; and hence the macromolecular weight is 304,000.

A somewhat different method is to determine the volume of a single $(\text{CH}_3)_2\text{SiO}$ unit and divide this into 520,000 Å³ to find the number of such units in the macromolecule. The dimensions of such a dimethyl siloxane unit have been estimated by Norton (3) to be 3.0 by 7.0 Å in surface area and 6.0 Å deep, and hence the volume is 126 Å³ (Norton's estimate is based on molecular models which assume covalent radii, and probably involve some error in the Si-O bond distance, according to the more recent ideas of Roth (7)). Dividing this into 520,000 Å³ gives 4120 such dimethylsiloxane units in the representative grain of Fig. 4, and since each dimethylsiloxane unit has a weight of 74.1 in atomic weight units, the molecular weight of our representative grain is 306,000, much the same as by the first method. By comparison with Scott's molecular weights in Table 1, it is seen that the structural units in the silicone matrix of Fig. 4 seem indeed to correspond to the real macromolecules.

Another estimate of the volume occupied by one $(\text{CH}_3)_2\text{SiO}$ unit may be made without recourse to models by utilizing the detailed and accurate crystallo-

graphic measurements reported by Roth and Harker (8) for octamethylspiropentasiloxane, $(\text{CH}_3)_8\text{Si}_5\text{O}_6$. This substance crystallizes in the space group D_{4h}^{19} ($I\ 4/AMD$), with $a = 14.09$ Å and $c = 10.18$ Å. There are four monomeric units (of the composition given above) in each unit cell, and the calculated density is 1.18. From the parameters the volume of the unit cell is 2016 Å³; from the density and the atomic weights the volume is 2030 Å³. If we consider each monomeric molecule as being composed of four $(\text{CH}_3)_2\text{SiO}$ groups and one SiO_2 group, then we have in the unit cell sixteen $(\text{CH}_3)_2\text{SiO}$ and four SiO_2 groups. The volume of an SiO_2 group may be estimated as about 2.2 Å³ in the particular structure; alternatively, we could use the volume of a silicon

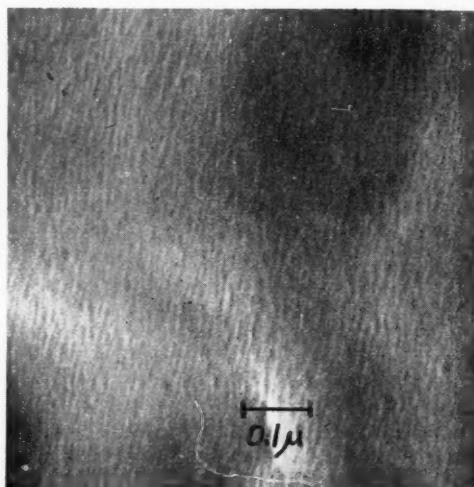


FIG. 5. Fracture surface of iron-red silicone rubber. Magnification $100,000 \times$.

tetrahedron in silicate minerals, which is about 1.5 Å³, for the volume is very small anyway in comparison with the organosilicon structures. Four SiO_2 groups therefore occupy about 6 Å³, leaving 2010 Å³ for the sixteen $(\text{CH}_3)_2\text{SiO}$ groups. This is equivalent to 126 Å³ per dimethylsiloxane unit, or exactly the same volume as Norton obtained, studying models.

It follows that the previous conclusion is supported, i.e., a particle or discontinuity 1.0 mm in diameter in Fig. 4 corresponds to a piece of dimethylsiloxane polymer of molecular weight 300,000, and hence is probably a single large molecule of the polymer.

More refined measurements were made on Fig. 5, which is a negative electron micrograph of a polyvinyl alcohol plus silica bireplica of a sample³ of "hard gum" silicone elastomer filled with relatively

³ By courtesy of A. Kneitel, Chemical Department, General Electric Company, New York City.

coarse material, presumably iron oxide. The filler represents little area in the picture and the macromolecules are abundantly portrayed. The smallest ones are round with diameter of about 100 Å and, calculated on the basis of specific gravity and Avogadro's number, their molecular weight is 310,000. Most of the particles in Fig. 5 are pictured elliptical: 100 Å wide and 200 Å long. Assuming 100 Å for the third diameter, the macromolecular weight is 610,000, which value also lies in the range given by Scott and listed in Table 1. The largest particles have dimensions roughly 100 Å by 800 Å. Assuming the third dimension to be also 100 Å, the macromolecular weight is 2,500,000, which is very close to Scott's maximum in Table 1.

It should be emphasized that measurements of the kind described herein require the highest resolution of which the RCA Model U electron microscope is capable, and further improvement in practical resolution is much to be desired. There may be many smaller molecules in Figs. 4 and 5, for example, than those which have been measured, and the shapes may appear to be rounded only because of their fuzzy outlines, which also render more precise measurements difficult. It is hoped that a new RCA accessory, the magnetic lens shield, placed around the path of electrons between the objective and projector lenses, will help produce much sharper outlines and consequently more precise measurements. Other improvements in technique will undoubtedly follow to refine the values to the extent that statistical descriptions and analyses may be obtained on the variations in sizes and shapes.

Then significant differences will be manifested among chemical and physical variations in the preparation of silicones and all other families of high polymers.

The distinct advantages of the resinographic method of studying the molecular structure of high polymers are:

1. It *pictures* rather than *infers* the shapes and sizes of the macromolecules.
2. The measurements are direct, the calculations simple, and the assumptions minor.
3. The method applies to all high polymers if they can be rendered brittle for fracturing and will remain rigid until a replica of the fractured surface is made. That is, the method applies to soluble high polymers whose molecular weights can be confirmed by established methods and it also applies to insoluble and infusible high polymers whose molecular weights have heretofore been unknown.
4. The method appears to be applicable to commercial (filled) plastics of fabricated shapes. The high polymeric matrix shows between the very distinctive particles of filler and other included material and there are always plenty of macromolecules of the high polymer for examination. Thus samples of special formulation, purity, or shape are not required.

5. The fracture surface is not destroyed by replication. Thus it is available for further experimental treatment such as aging, curing, or other influences on molecular sizes and shapes.

The authors gratefully acknowledge the assistance of F. G. Rowe and M. C. Botty in preparing the electron micrographs used in this article.

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Technical Papers

Ovular Tumors and Inhibition of Embryo Growth in Incompatible Crosses of *Datura*¹

J. Rappaport, S. Satina, and A. F. Blakeslee

Smith College Genetics Experiment Station,
Northampton, Massachusetts

For some time the Smith College Genetics Experiment Station has been making a somewhat detailed study of the various barriers to crossability among the 10 herbaceous species of the genus *Datura*. Of the 90 interspecific combinations, with each species used both as a male and as a female parent, only 19 have resulted in viable

tegument outside the endothelium are at first filled with starch grains. With the growth of the embryo this starch is depleted. As the endosperm develops inside the embryo sac it becomes filled with fat and aleurone grains. The endothelium remains a single layer of cells which lose their contents and become much flattened. Ultimately the layers outside the embryo sac, including the endothelium itself, become digested.

In incompatible crosses the condition in the young ovule is similar to that just described for compatible crosses. Soon, however, the cells of the endothelium stain more deeply and enlarge, as shown in Fig. 2. The outlying cells of the integument do not lose their starch. Chemical tests seem to show that the ovular contents of selfs and compatible crosses convert starch (presumably

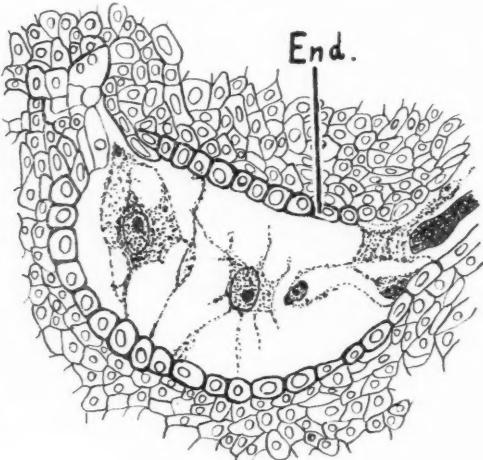


FIG. 1

hybrid seed. In some cases the pollen tubes burst in the style of the foreign species or grow too slowly to reach the ovary. It seems to be the rule, however, that if the sperm can be gotten into contact with the egg cell, fertilization results. The postfertilization barriers to crossability which result in abortion of the hybrid embryos can be overcome in many crosses by cultivating the excised embryos on artificial media before they abort.

There are great differences in the behavior of tissues within the ovules of incompatible and compatible crosses or selfs. In compatible crosses and selfs, a single layer of cells, the endothelium (marked "End." in Fig. 1), which is epidermal in origin, surrounds the embryo sac and these cells act as nurse cells in passing on nourishment to the developing embryo. The cells of the in-

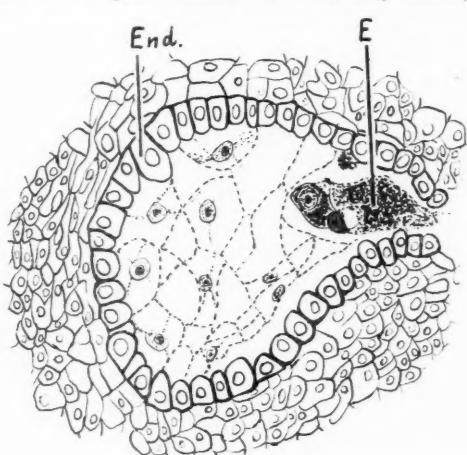


FIG. 2

due to an amylolytic enzyme), whereas the contents of ovules from an incompatible cross are apparently unable to convert starch. It would seem, therefore, that in incompatible crosses either an amylolytic enzyme is absent or its activity is inhibited. Accompanying the growth in size and division of the endothelium in all planes, there is abortion of the contents of the embryo sac. Sometimes the endosperm aborts first and sometimes the embryo, as is the case in Fig. 2 (the embryo is marked "E"). In more advanced stages, such as that shown in Fig. 3, the endothelial tissue may proliferate at several points and penetrate into the embryo sac to form groups of tumoral tissue. Sometimes such ovular tumors form definite masses visible to the naked eye, which from their appearance have been called "pseudo embryos."

The contents of ovules from incompatible crosses may be watery, jellylike, milky, or cheeselike, dependent somewhat upon which parents had taken part in the cross and upon the age of the ovule. It should be

¹Contribution from the Department of Botany, Smith College, New Series No. 41. This research was supported in part by the Committee on Growth of the National Research Council acting for the American Cancer Society and by the U. S. Public Health Service.

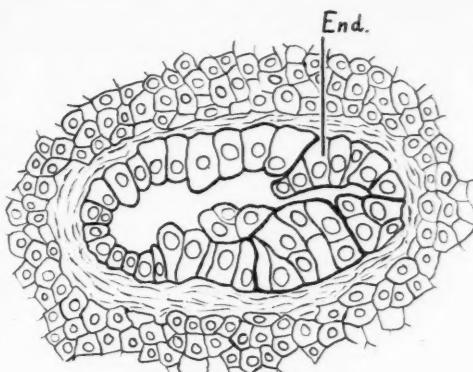


FIG. 3

pointed out that the stimulus to incompatibility and ovular-tumor formation is not confined to species crosses in which qualitative differences in the genes of the two parents might be supposed to be in some way responsible for the failure of embryo growth. Ovular tumors and incompatibility occur also in $4n \times 2n$ crosses within inbred lines of the same species.

Ovular tumors associated with embryo abortion in incompatible crosses of *Datura* have been found to contain a water-soluble thermostable substance unrelated to plant hormones, which is capable of inhibiting growth of embryos from selfed *D. stramonium* both *in vitro* and *in vivo*. The contents of such inhibited ovules contain a substance capable of inhibiting another set of capsules. This substance has been found effective in three successive passages, suggesting a self-duplication such as occurs in viruses or a new formation of inhibitor stimulated by the originally injected ovular-tumor extracts. Fig. 4 diagrams the effects of these injections.

In the upper part of the diagram is shown that one-fourth of the extract from a single inhibited ovule (a) will not inhibit ovules in another capsule, whereas the extract from a whole incompatible ovule (A) inhibits the

± 100 ovules of the locule of the capsule into which it has been injected. The extract (B) from one of these ± 100 inhibited ovules will contain one-hundredth of the amount of inhibitor present in A. Since, however, it inhibits a second series of ± 100 ovules it is believed that the strength of the inhibitor had in some way been increased. The strength of the extract (D) which inhibits ovules in capsule IV must be only one-millionth of that in the original (A) if no such increase had taken place.

The basis of this increase in potency of the inhibiting substance, as well as the nature of the inhibition, is receiving further study.

Solution of this problem might well lead to methods whereby the postfertilization barriers to crossability could be removed, with a great increase resulting in the number of wide species hybrids possible. It might also throw new light on the broader problems of both normal and abnormal growth and differentiation in other forms.

Details of our studies on ovular tumors in *Datura*, together with literature citations, will be given in two papers now in press in the *American Journal of Botany*.

A Mechanical Heart with Coagulable Blood

Lucien Brull

*Institut de Clinique et de Polyclinique Médicales,
Université de Liège, Belgium*

As far as we know, no artificial heart with coagulable blood has yet been devised. Our method, which will be described briefly, aims to take blood (arterial, venous, or mixed) without extravasation, from any part of the body, raise or lower its pressure to any wanted level, and perfuse any organ, organs, or part of the body with this blood, which is sent back to the right heart through a jugular vein.

This artificial heart is made from an aorta in which the blood is propelled by a roller-pulley. The peripheral resistance is provided, on the one hand, by the perfused organ or organs, and on the other, by a shunt such as we described previously (1), which is coated inside by a carotid. The air pressure exerted on this vessel regulates the arterial pressure.

The aorta of a dog of 15–30 kg is dissected from the heart up to and including the iliac division, and all branches carefully ligatured. It is cut 5 cm below the subclavian artery. The central end of the peripheral part is turned inside out over a Payr's cannula of 10–12 mm diam. The cannula is tied at its other end inside the end of a piece of rubber tubing of 12-mm diam and 80-mm length. The distal part of the vessel emerges from the rubber tube. The superior mesenteric branch is connected with the shunt, which is the pressure regulator. One of the renal arteries is connected with a manometer. One of the iliac arteries leads to the perfused organ or organs (i.e., kidneys); the other iliac artery, clamped, is used for rinsing the air out of the preparation.

The crook of the aorta, which has been separated, is tied at its peripheral end over the central end of the other part of the aorta, which is fixed on the Payr's cannula.

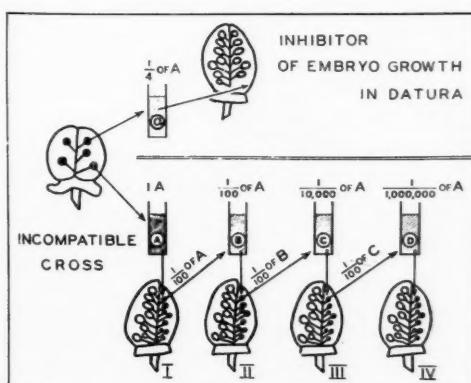


FIG. 4

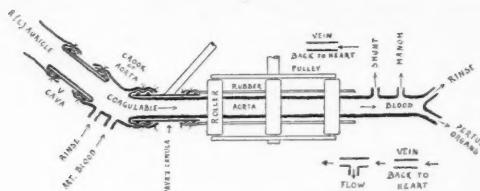


FIG. 1. Artificial heart with coagulable blood.

The subelavian artery is connected with a reservoir of saline or blood to rinse the air out of the preparation. The innominate is either tied or connected with an artery to introduce arterial blood into the preparation. Into the large end of the aorta, a Payr's cannula of 15-mm diam, coated with a vena cava, is introduced and tied. The other end of this cannula is introduced into the auricle, right or left, according to which sort of blood one wants to pump out of the heart.

The rubber tubing which protects the aorta rests by means of a layer of moss rubber on a flat groove. The roller-pulley can be raised or lowered by a screw so that the rollers as they rotate exert more or less pressure on the aorta (systolic output), and the speed of the pulley can be regulated to vary the number of beats. The diameter of the pulley is 65 mm and there are six rollers of 8-mm diam. In order that the aorta may not slip off, the cannula on which it is tied is itself fixed on the apparatus.

The output of such a heart can easily reach 700-1000 ml/min, against a pressure ranging from the normal level up to 300 mm. Up to the present it has been used to perfuse kidneys with arterial blood under varying pressures, or with venous blood at arterial pressure. Brief results of these experiments will be published at the International Congress of Physiology in 1950.

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Hollow Crystals of Nitroguanidine¹

Joseph Cohen, Ronald A. Henry, Sol Skolnik, and G. B. L. Smith²

*Inorganic Chemistry Section, Chemistry Division,
U. S. Naval Ordnance Test Station,
China Lake, California*

The irregular formation and occurrence of voids and internal imperfections in crystals have been observed frequently. However, the consistent and reproducible formation of regular-shaped cavities along the entire length of a crystal has not been previously observed, to our best knowledge. We have found recently that under certain conditions nitroguanidine crystallizes from solution as

¹ Published with the approval of the Chief of the Bureau of Ordnance, U. S. Navy.

² The authors wish to acknowledge the assistance of Messrs. Don MacLaghlan and Al Uremovich, who prepared the thin sections and the photomicrographs.



FIG. 1. Photomicrograph of nitroguanidine.

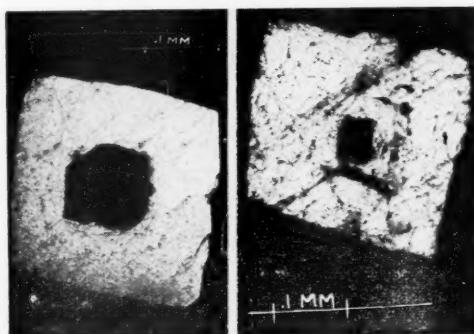


FIG. 2. Photomicrographs of thin cross sections of hollow nitroguanidine crystals.

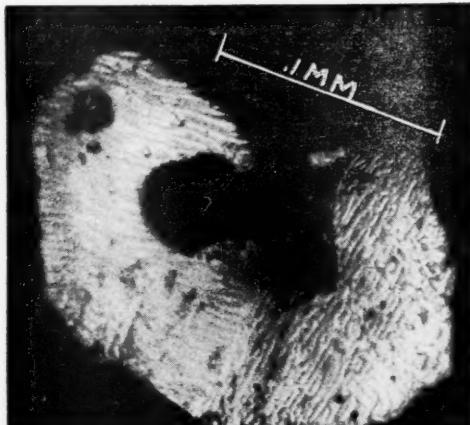


FIG. 3. Photomicrograph of a thin cross section of an incompletely formed hollow nitroguanidine crystal.

long, hollow, flexible needles (Fig. 1). The solvent used is a 1.2% aqueous solution of acetic acid; the concentration of nitroguanidine is 5 g/l, and the cooling time, from 95° C to 30° C is 5 hr, with no agitation. The resulting needles are not single crystals but appear to be aggregates of crystals.

Photomicrographs of cross sections made by embedding the needles in plaster of Paris or very hard wax and grinding thin sections perpendicular to the long geometric axis of the needle are shown in Fig. 2. From a study of a large number of cross sections it appears that the boundaries of the cavity are always parallel to the external faces of the aggregate, and that the respective numbers of faces are the same. That is, if the cross section of the aggregate is hexagonal, the cavity will be hexagonal; or, if the cross section is square, the cavity will also be square. A photomicrograph of the cross section of an incompletely formed hollow aggregate is shown in Fig. 3.

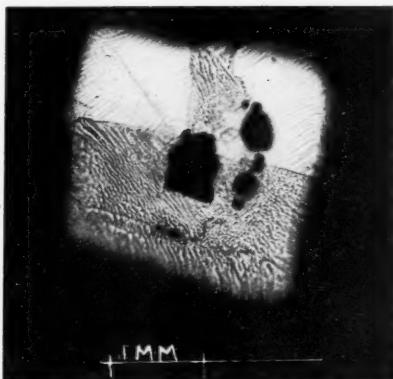


FIG. 4. Photomicrograph of the same cross section as shown in Fig. 2 on right, but after etching with water.

The presence of the cavity throughout the length of the aggregate is easily demonstrated in another way. If one end of the aggregate is dipped into an aqueous dye solution, the colored solution can be seen to ascend the bore of the aggregate, due to capillarity. Under the microscope a well-developed meniscus is readily seen. Frequently, the ends of the cavity are partially or completely closed because of crystallization, in which case it is necessary to trim the ends of the aggregate before the capillary can be revealed.

In any given crystallization under the conditions described, approximately 70%–80% of the needles will have well-defined cavities. If the crystallization is conducted from water rather than dilute acetic acid, the percentage of hollow aggregates is small (10%–15%). The influence of concentration or of the rate of cooling on this phenomenon has not been investigated. No effort was made to ascertain the minimum crystal size that still possesses the cavity; however, some aggregates with diameters as small as 0.2 mm have been hollow.

The formation of these hollow aggregates is probably due to a lengthwise contact twinning, involving two or

three crystals. If the cross sections are carefully etched with water, groups of parallel striations appear, which are independent of the direction of grinding, and which appear to define the individual crystals composing the hollow needle (Fig. 4).

Cursory examination indicates that some of the long flexible needles of sublimed phthalic anhydride are also hollow.

Vacuum Differential Thermal Analysis¹

Walter L. Whitehead and Irving A. Breger²

Department of Geology, Massachusetts Institute of Technology, Cambridge

Differential thermal analysis is a technique for studying certain thermal characteristics usually of a solid complex substance. The procedure consists of heating the material under investigation at a constant rate to about 1000° C and recording the temperatures at which exothermic and endothermic reactions occur. In practice this is most readily accomplished by employing a metallic block with two holes, one of which is packed with the unknown substance, while the other is packed with an inert powder such as γ -alumina. A two-headed thermocouple, one head of which is imbedded in each powder, is then used to determine the direction and extent of the differential changes as the block is heated at a constant rate in a furnace.

During recent years differential thermal analysis has become a standard analytical procedure for the study of complex mixtures of minerals when resolution is difficult by conventional microscopic techniques or by x-ray methods. It has been especially useful in analysis of clays and other colloidal materials.

During the course of studies on coals and organic shales it became evident that any simple analysis or set of analyses leads only to generalized indication of the structure and stability of the gross organic material. Difficulty is enhanced by the fact that these organic substances, derived to a great extent from colloidal gels, appear homogeneous under the microscope. Differential thermal analysis has proved after trial (1–3) to be a promising technique for such studies, but certain difficulties were encountered in determining the thermal characteristics of the organic complexes on account of combustion of the specimens. For this reason a new instrument for differential thermal analysis was designed to operate under vacuum or inert atmosphere. This paper describes such a vacuum installation, its furnace, and control equipment. A typical record is illustrated in Fig. 1.

Previous differential thermal analytical studies have been carried out with magnesia-packed furnaces, but the

¹ The research on marine shales was sponsored by the American Petroleum Institute, and that on coals by the Nova Scotia Research Foundation.

² The authors wish to acknowledge suggestions by Mr. Coutland Pearsall on furnace design, and the cooperation of Mr. John Solo in construction of the furnace.

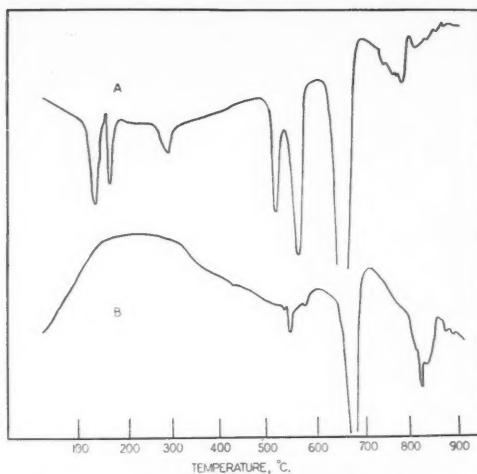


FIG. 1. Thermographic record of (A) pyritized coal from Greene County, Indiana (100 mg) and (B) pyrite (25 mg).

high thermal capacity of the packing has made it difficult to attain heating rates exceeding 10° or 12° C per min. For this reason, and because of the impracticality of attempting to evacuate a packed furnace, a vertical heating unit was designed with radiation shielding rather than conventional packing.

The furnace (Fig. 2) is constructed with a specially

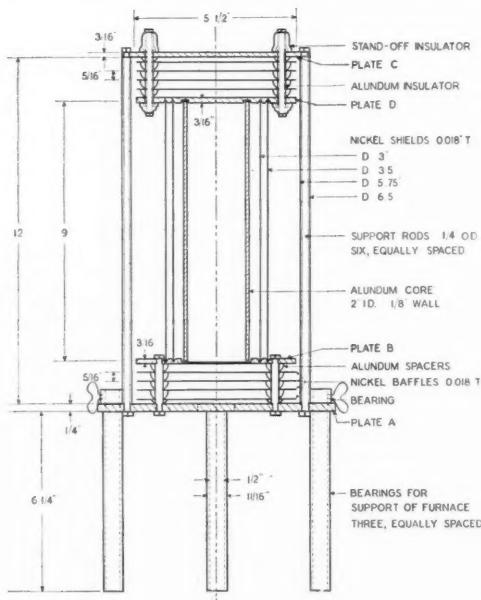


FIG. 2. Furnace used for thermographic analysis showing arrangement of radiation shields.

cast Alundum core 9 in. long, 2 in. inside diam, with a $\frac{1}{8}$ in. wall.³

Sufficient 16-gauge Chromel-A wire is wound onto the core to make the power input approximately 2 kw at 110 v a-c. The core is shielded by four cylinders constructed of 0.018-in. sheet nickel. On top and bottom, shielding is by four disks made of the same material. Electrical connections within the furnace are made with stainless steel lugs and 1-mm platinum wire. For electrical insulation ceramic fish-spine beads are strung over the leads.

All steel parts of the furnace are machined from material obtained from the Rustless Iron and Steel Corporation, Baltimore, Maryland. Steel type 446, containing 23.00% to 30.00% chromium and a maximum of 1% nickel, and type 309, containing 22.00% to 26.00% chromium and 12.00% to 14.00% nickel, are used interchangeably, depending upon availability. These steels were chosen because of their resistance to sealing and to sulfur at high temperatures.

The furnace is mounted on three posts, 18 in. long, screwed into an 18 in. \times 18 in. \times $\frac{1}{2}$ in. brass plate. A groove $\frac{1}{2}$ in. deep and of width to accommodate an inverted 12 in. \times 24 in. Pyrex jar is machined into the plate. To prevent failure of the glass jar, its flat bottom is rounded and then the entire jar is carefully annealed. By setting the plate on 2-in. adjustable legs and screwing and silver soldering a brass line into a hole through the plate, a means is afforded for complete evacuation of jar and furnace or for replacement of the air by an inert atmosphere such as helium. Electrical leads to the furnace and all thermocouple leads are insulated from the base plate by means of Kovar-to-glass seals obtained from the Stupakoff Ceramics and Manufacturing Company, Latrobe, Pennsylvania.

The block (Fig. 3) in which analysis is carried out is machined from type 446 or type 309 steel, has a diameter of 1 in. and has a length of $\frac{1}{4}$ in. Its lower end has a groove $\frac{1}{2}$ in. deep to enable the block to set firmly in place on a ceramic 1 in. outside diam \times 12 in. furnace core which serves as a support post. This post is firmly fixed onto the base plate by means of a tightly fitting brass sleeve. The furnace bearings are of such length that when they rest on the brass plate, the heating block is exactly centered in the furnace.

The heating rate of the furnace is controlled from a Chromel-Alumel thermocouple junction placed between the center two windings of the spiral heating element. The output of the thermocouple is fed into a Leeds and Northrop Micromax controller equipped with an auxiliary device which enables variation of the heating rate of the furnace from 1° to 50° C per min. A heating rate of 12° C per min has been chosen for all comparative work in this laboratory. Additional control devices enable a constant cooling rate and "soak" of a specimen at a predetermined temperature.

Block temperature is measured by a Chromel-Alumel junction placed in the center hole of the block. The output of the thermocouple is recorded on a Brown recorder.

³ Prof. F. Vinal of the M.I.T. Ceramics Division was instrumental in obtaining this core.

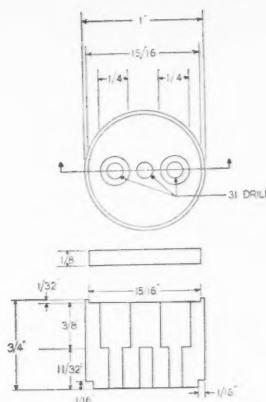


FIG. 3. Sample block.

The differential thermocouple is made from two leads of 28-gage Chromel wire connected by a short length of 28-gage Alumel wire. The fine wire is used in order to eliminate conduction of heat away from the specimen and the short length of connecting Alumel wire is used for the same reason. The output of the differential thermocouple is fed through a resistance box into a galvanometer having a short time constant. The position of a light beam reflected from the mirror of the galvanometer suspension is recorded on a Beckman Photocell Recorder (National Technical Laboratories, Pasadena, California).

For automatic operation of the installation, the control unit, recorders, and galvanometer light are wired through a Type T-27 General Electric time switch which may be adjusted to start the run and then to turn off the installation (see Fig. 4) after a preset time interval.

Samples of 25 mg to 100 mg of material, ground to pass a 200-gage screen, are being used for the analysis. For maximum effectiveness the samples are packed immediately around the thermocouple junction. Using specially designed tools, it is possible to pack the material consistently into the sample hole at 530 psi, while at the

same time assuring horizontal and vertical centering of the thermocouple junction.

The sensitivity of the differential recording installation is approximately ± 0.15 millivolt. This sensitivity can be increased by the substitution of a more sensitive galvanometer, a procedure that should enable the analysis of samples of smaller size.

During current operating practice with organic materials, the bell jar is evacuated to 1 mm of Hg and pump operation is continued throughout the experiment. The analysis of clays or inorganic samples can be made without the use of vacuum.

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Developmental Failure of the Pituitary in Amphibian Embryos Treated with Sugar

Richard M. Eakin

Department of Zoology, University of California, Berkeley

A striking syndrome of abnormalities was obtained in embryos and larvae of the Pacific tree toad, *Hyla regilla*, by immersing late blastulae or beginning gastrulae, unremoved from the jelly, in a 10% solution of sugar (sucrose) for 14-20 hr at room temperature. Gastrulae so treated showed retardation of early gastrular movements and complete inhibition of gastrulation beyond the large yolk-plug stage. An exposure longer than 24 hr was usually lethal. Neurulae, developing from gastrulae returned to pond water, exhibited varying degrees of foreshortening of the archenteron and, correspondingly, of the medullary plate. Abnormalities frequently observed in the tail-bud stage included delayed formation and reduced size of the stomodeum, partial or complete fusion of nasal and sucker placodes, small irregular optic vesicles, and dorsal bending of the body. Larvae were characterized by the following features: albinism of the type caused by a deficiency of the melanosome-expanding hormone of the intermediate lobe of the pituitary; monorhina; reduction or complete absence of mouth parts; fused suckers; reduction in size and irregularities in form of eyes; partial or complete *situs inversus* of the gut; circular swimming movements owing to the dorsal bending of the body and tail; and little or no progress toward metamorphosis. All of the above abnormalities appeared in various combinations and to different degrees according to the length of treatment. Larvae developing from embryos immersed in the sugar solution for 14-16 hr showed mild symptoms; those exposed for 18-20 hr were highly abnormal. These observations have been made repeatedly and upon large numbers of animals. The cause of the anomalies has not yet been analyzed, but it would appear that because of the disturbance to gastrular movements—presumably an osmotic effect—the inductive role of the mesoderm was impaired,

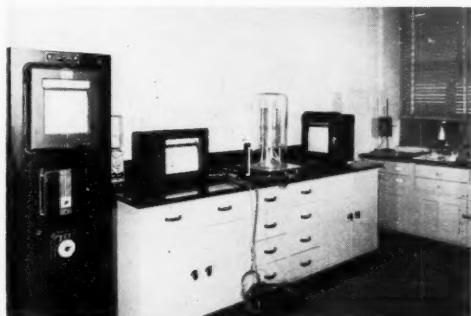


FIG. 4. Photograph of installation for thermographic analysis showing furnace, controller, recorder, galvanometer, time switch, vacuum gage, and pump.

possibly through altered spatial and temporal relationships between the mesoderm and ectoderm or possibly through a direct physiological effect of the treatment upon the mesoderm.

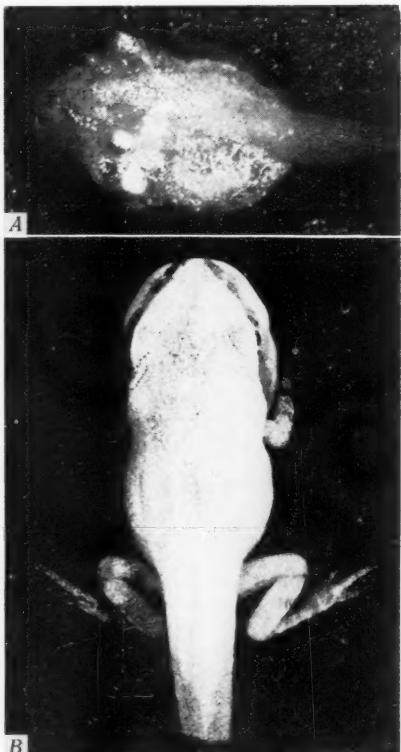


FIG. 1. Larvae of *Hyla regilla* developing from gastrulae treated with sugar. A. Young tadpole exhibiting albinism, monorhina, and abnormal eyes. B. Thyroid-fed albino tadpole in a final stage of metamorphosis.

In this preliminary report only one aspect of the picture will be given further comment, namely, the hypopituitary effects. Albinism appeared in a high percentage of the larvae developing from gastrulae treated for 16-20 hr. The silvery appearance of the tadpoles (see Fig. 1A) was identical with that obtained by hypophyseotomy by Smith (5), Allen (1), Burch (4), and others. Both dermal and epidermal melanophores were markedly reduced in number, and their melanin pigment was highly concentrated so that the melanosomes appeared as dots. The xanthophores, on the other hand, were numerous and their yellow pigment fully dispersed. In older animals the xanthophores formed a continuous silvery sheet over the body and dorsum of the tail. The eyes became so covered with xanthophores that the black tapetum was almost entirely obscured.

The second manifestation of hypophyseal deficiency was poor progress toward metamorphosis. Ten or twelve

experimental animals were successfully maintained until the controls, developing under identical conditions, had completely metamorphosed. These animals—albinos, of course—were entirely larval in character except for the development of small limb buds. A few specimens attained a differentiation of the hind limb represented by stage XI of Taylor and Koilros (6), in which the rudiments of all five digits are present. Mouth, skin, gut, and other structures, however, showed no metamorphic changes. The tadpoles grew in size, nevertheless, some exceeding the controls in bodily length. To a group of six large albino larvae, thyroid substance was fed. In 14-20 days, four of the surviving animals metamorphosed. One of the young frogs in a final stage of metamorphosis is shown in Fig. 1B. Even after metamorphosis the skin was completely silvery, owing to the very numerous and fully expanded epidermal xanthophores. In certain regions only, such as in the larval skin of the tail (see bottom of Fig. 1B), could the melanophores—still highly contracted—be observed.

The cause of these manifestations of hypopituitary function lies in the failure of the pituitary to develop normally. In all of the animals, about twenty thus far sectioned and examined microscopically, the infundibulum was poorly formed, and the hypophysis was a small, single body of cells lying beneath, but not in contact with, the brain. In some instances the hypophysis was separated from the brain by cartilage, in others by connective tissue. In no specimen could lobes—*paries distalis* (anterior), *intermedia*, and *tuberalis*—be observed. Thus the hypophyses was morphologically undifferentiated. The hypophysis of young larvae seemed also undifferentiated histologically, but those of older albinos exhibited some differentiation, such as acidophils. As yet, however, only hematoxylin-eosin preparations have been made.

The consistent picture of albinism, even after induced metamorphosis, and the absence of a *paries intermedia* in these sugar-treated animals, give added support to the theory of Blount (2, 3) and Burch (4) that the differentiation of the intermediate lobe of the pituitary requires contact of the hypophysis with the floor of the brain, perhaps specifically with the infundibulum.

That a similar conclusion—supported by Burch but denied by Blount—holds for the *paries distalis* is not clear in these experiments. The fact that my sugar-treated animals showed only poor progress toward metamorphosis until they were fed thyroid substance points to a deficiency of thyrotropic hormone resulting from a defective development of the anterior lobe. In this connection it should be noted that Blount's albino *Ambystoma* larvae did not metamorphose, and his photomicrographs (3, Figs. 14 and 19) of their thyroids show follicles with dense colloid and low follicular epithelium, suggestive of inactivity. Yet he infers the presence of an active anterior lobe. On the other hand, in my sugar-treated animals there was some evidence of histological differentiation in the hypophysis, some progress in the development of limbs, and in some specimens a definite picture of thyroid activity. These facts speak against a claim for dependent differentiation of the *paries distalis*.

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A Very Water-soluble Riboflavin Derivative

George B. Stone

Technical Service Department, Chas. Pfizer & Co., Inc., Brooklyn, New York

For the past ten years a not inconsiderable amount of investigation has been carried out in an effort to increase the water solubility of riboflavin (vitamin B₂) either by the use of solubilizers or by the preparation of soluble derivatives. Despite some thirty or more references and patents, representing several hundred solubilizers or soluble derivatives, few, if any, are of practical significance for pharmaceutical application. Riboflavin is not only sparingly soluble in water, but in almost every other solvent. It is, however, relatively soluble in concentrated sulfuric acid. Investigation of this significant solubility in concentrated sulfuric acid led to the isolation of a very water-soluble riboflavin derivative.

The compound was prepared by dissolving 50 g of riboflavin, little by little, in 200 ml of concentrated sulfuric acid with vigorous stirring, while the temperature was maintained at 40°–50° C. Mixing was continued for 1–2 hr until the mixture was homogeneous and it was then quenched by pouring it over 1 kg of cracked ice. The resulting solution was neutralized with slurried calcium hydroxide to a pH of 6.5, with the temperature being maintained below 70° C. The precipitated gypsum was filtered off, washed with hot water, and then repulped with hot water, filtered, and again washed. All washings were added to the original filtrate. Assay by the fluorometric method indicated that the original 50 g of riboflavin was present in this solution. The solution was concentrated under vacuum to a volume of less than 500 ml and filtered to remove further gypsum precipitated during concentration. The filtrate was then freeze-dried to yield 114 g of a fluffy yellow-orange powder, which assayed fluorometrically 57.2% riboflavin, equivalent to a yield of 100% based on the weight of the riboflavin employed originally.

The compound is stable in air and nonhygroscopic. It is very soluble in water, and aqueous solutions containing 10% wt/vol of riboflavin have been prepared—a solubility 1,000 times greater than that of riboflavin U.S.P. It is soluble in methanol and slightly soluble in ethanol, and it decreases in solubility with the higher alcohols. It is soluble in glycerine, propylene glycol, and pyridine; slightly soluble in acetone, glacial acetic acid, and chloroform; insoluble in benzene, ether, ethyl acetate, methyl-ethyl-ketone, and carbon tetrachloride. Aqueous solutions are heat-stable at 15 psi for 120 min in the pH range from 1.0 to 6.5.

Preliminary chemical analysis seems to indicate that the compound may be represented by the empirical formula, C₁₇H₁₈N₄O₅S₂Ca, inasmuch as the compound contains calcium and sulfur, a portion of the sulfur being present as sulfate.

Fluorometric assay of the material yields a value of 57.2% riboflavin. The absorption spectrum is identical with riboflavin U.S.P., having the same maxima and minima, but proportionately displaced because of the lesser riboflavin content. Paper chromatographic absorption analysis indicates the material is a pure compound, much more water-soluble than riboflavin U.S.P.

Microbiological assay by the U.S.P. XIII revision method, which includes a preliminary hydrolysis at 15 psi for 30 min, yielded a value of 33.0% riboflavin. Omission of the preliminary hydrolysis gave a value of 1.5% riboflavin, whereas increase of the time of hydrolysis to 120 min gave a value of 42.2% riboflavin.

Biological assay for riboflavin by a standard rat growth method employing a basal vitamin B complex-free diet, supplemented with those members of the B complex other than riboflavin, indicated that the riboflavin potency for the rat is almost nil. There were indications of a slight antivitamin activity of this compound.

Further investigation of this material is anticipated.

A Correction for Linkage in the Computation of Number of Gene Differences

Everett R. Dempster and Leon A. Snyder¹

Genetics Division, University of California, Berkeley

The Castle-Wright formula (2) for estimating the number of gene pairs differentiating two strains with respect to some quantitative character is based on the increased variance of the F₂ as compared to the parental variance. A number of postulates on which this derivation is based (1, 5, 6, 7) may be listed as follows: (1) The parents are homozygous. (2) All the plus alleles differentiating the two strains with respect to the character considered are in one parent and all the minus alleles in the other. (3) All gene differences affecting the character have equal effects. (4) The effects of different allelic substitutions are additive. (5) There is no linkage. Deviations from postulates 2 to 5, with the possible exception of special epistatic effects (postulate 4) will always increase the F₂ variance, and therefore, since the latter appears in the denominator of the expression for gene number, will bias the estimate toward lower values. Since actual situations are usually at variance with most of the postulates listed, the expression in general leads to minimum rather than unbiased estimates of the number of gene differences.

Some modifications have been devised for relaxing the postulates or otherwise extending the applicability of

¹ Present address: Department of Plant Sciences, University of Oklahoma, Norman, Oklahoma.

gene number estimates based on variance. Thus Serebrovsky (6) has presented formulas utilizing backcross data in which allowance is made for any degree of dominance, and Charles and Goodwin (3) have considered the extension of the method to the case of several correlated characters. The purpose of this note is to suggest a correction for linkage.

The formula given in the article by Castle (2) is $n = D^2/[8(\sigma_e^2 - \sigma_1^2)]$ where D is the difference between the means of the parental races, σ_1 is the standard deviation of the F_1 generation, and σ_e is the standard deviation of the F_2 generation. The difference term in the denominator, which will be written σ_e^2 in the following discussion, represents an estimate of that portion of the F_2 variance due to segregation of genes at loci for which the parents were homozygous for different alleles.

It has been shown (4) that if n loci, at which nondominant alleles have equal differential effects, are distributed at random among m linkage groups within which linkage is complete, the expected factor of increase in genetic variance due to linkage is $(m+n-1)/m$. If we divide σ_e^2 by this factor, the quotient is an estimate of the genetic variance that would be obtained in the absence of linkage. We have then, for the case of complete linkage, the expression $n = [(m+n-1)D^2]/[8m\sigma_e^2]$, from which can be obtained the following:

$$n = [m-1]/[(8m\sigma_e^2/D^2) - 1].$$

For comparison the uncorrected formula may be written:

$$1/[8\sigma_e^2/D^2].$$

Since real chromosomes contain unequal numbers of genes and are subject to crossing over, their number cannot be directly substituted for m in the above formulas. Instead one should substitute the number of hypothetical chromosomes, possessing equal proportions of the total number of genes and immune to crossing over, that would increase F_2 variance to the same degree as do the real chromosomes. The following considerations lead to an approximation for m which is conservative in the sense that it somewhat underestimates the average effect that linkage would be expected to have in increasing variance. If there are k points of exchange in a bivalent chromosome, there are $(k+1)$ terminal and interstitial regions. At any exchange point two homologous chromatids are involved, the other two remaining unaltered. In the absence of chromatid interference, any of the $2^{(k+1)}$ possible combinations of the $(k+1)$ regions, with respect to origin from one or the other homolog, is equally likely to appear in any given haploid nucleus resulting from meiosis. This is equivalent, with respect to F_2 variance, to the independent assortment of $(k+1)$ pairs of chromosomes within which linkage is complete, although the lengths of the hypothetical chromosomes would be unequal. The expected effects of linkage on F_2 variance can readily be shown to be greater for chromosomes of unequal as compared to chromosomes of equal length (4). The over-all effect on linkage, therefore, approximates and is somewhat greater than that which would result from the division of the chromosome complement into $\Sigma_i c_i (k+1)$ equal segments within which linkage

is complete, where c_i is the actual number of chromosome pairs and k the average number of exchanges in each pair. Since fifty crossover units correspond to an average of one exchange chiasma per bivalent, the sum of one-fiftieth of the total number of map units and the number of chromosome pairs may also be employed as a rough estimate of the effective value of m . However, chromosomes that are very small compared to the others in the complement and much less than 25 map units in length would contribute very little to expected increase of variance and should be excluded in computing these approximations.

The value of m for maize on the basis last mentioned would be at least 27, and for *Drosophila melanogaster*, excluding the tiny fourth chromosome, approximately 9. There is, however, no crossing over in male gametogenesis in *Drosophila*, and so a more reasonable value for m in this organism would be the mean of the value just given, 9, and the number of major chromosome pairs, 3 or 6. Sex linkage would probably somewhat increase the variance among F_2 males and decrease it among F_2 females.

It may first be noted that the corrected formula leads to greatly increased estimates where the variance values are such that the uncorrected formula would indicate around m or more loci. Thus in *Drosophila melanogaster*, if m is taken as 6, some values of n given by the uncorrected and corrected formulas, respectively, are as follows: 3 and 5, 5 and 25, 6 and ∞ .

The error variance of σ_e^2 is the sum of the error variances of σ_1^2 and σ_e^2 and is not likely to decrease much with increase in gene number. Therefore the accuracy with which estimates may be made decreases very rapidly as gene number increases, whether there is independent assortment or linkage. An error variance just small enough for a distinction to be made between 3 and 4 loci would barely permit discrimination between 6 and 12, or 9 and 30, or 12 and ∞ loci.

Any error in the estimated value of σ_e^2 will contribute slightly more to the inaccuracy of gene number determinations with linkage than would be the case with independent assortment. A measure of the relative effects due to this cause may be obtained by comparing $dn/[d(\sigma_e^2/D^2)]$ for the two formulas. The two values are $8n^2$ and $8n^2[m/(m-1)]$ for independent assortment and linkage respectively. Linkage thus increases the standard error by the proportion $1/(m-1)$. This increase refers, however, to a component of error that could be minimized to any desired degree by taking sufficiently large samples. Another and probably much more important effect of linkage on error is dependent on the actual distribution of loci among linkage groups in the particular parental strains considered. For any particular pair of parental strains, the loci involved would usually be either more or less uniformly distributed among the different linkage groups than mean expectation. This would be true, in general, even in the absence of special mechanisms, such as the introgression of genes from one species to another, that might favor a concentration of genetic differences in one or a few linkage groups. Thus in any particular case there may well be a serious bias in the plus or minus direction of gene number estimate, and

such a bias cannot be reduced by an increase of sample size.

In conclusion, then, it is possible to apply a simple correction for linkage to the formula for estimating the number of different genes. This correction, if the postulates on which it is based are met, applies to the average situation. In the case of any particular pair of parental strains, the actual distribution of differentiating loci on linkage groups may lead to a serious bias in estimates of gene number. Even in the absence of the complications due to linkage, the estimate of gene number is likely to be highly inaccurate except where the number of differentiating loci is relatively small.

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Correlation of Certain Physical Constants of Some Alkyl Esters of *n*-Phenyl Carbamic Acid, with Their Phytotoxicity¹

V. H. Freed

Oregon Agricultural Experiment Station,
Corvallis, Oregon

The toxicity of the alkyl ester derivatives of phenyl carbamic acids to members of the Gramineae family was first demonstrated in 1929 by Freisen (4), dealing with ethyl *n*-phenyl carbamate. Deysson (2) in 1945 demonstrated that this material acted, at least in part, as a mitotic poison, inhibiting cell division and causing subsequent death of the cell. This was a direct corroboration of the work done by Lefevre (5) in 1939.

Templeman and Sexton (10) in 1945 reported on the phytotoxicity of various carbamates, with the object of controlling certain weedy plants. In 1946 the same authors (11) announced their discovery of the phytotoxicity of isopropyl *n*-phenyl carbamate. Allard *et al.* (1) in 1946 further elaborated on this phytotoxicity. Numerous investigators have since published information concerning the merits of this chemical as a phytocide.

There have been many attempts to correlate chemical structure with the biological activity of a number of compounds. Frear *et al.* (3) undertook the study of some 5,000 organic chemicals to elucidate certain chemical structures which could be correlated with toxicity. They concluded that certain groupings in specific types of aporadicals produce a toxic entity. Tattersfield and Roberts (9) reported a study of physical properties and

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TABLE 1

Alkyl group	Bp(Alc) °C	MP (deriv.) °C	Density (deriv.)	Refractive index (91° C)
Methyl	66	47	1.15	1.5235
Ethyl	78	52	0.92	1.5105
Propyl	97	58	1.06	1.5056
Isopropyl	83	90	1.09	1.4989
Butyl	116	57	1.03	1.4987
Sec-butyl	99	30	1.70	1.4957
Isobutyl	108	80	1.60	1.4955
Amyl	138	46	1.01	1.4926
Isoamyl	130	55	0.98	1.4939

chemical constitution of organic compounds as related to their toxicity to the wireworm. These authors concluded that in any homologous series the most toxic compound would be the one with the highest vapor pressure if it possessed a sufficiently high molecular weight. Rubbo (7) demonstrated the correlation of the ionization constant to toxicity for mice and bacteria of derivatives in the acridine series.

Melander (6) was able to correlate the physical constants of various isomers of hexachloreyclohexane with their toxicity. He found that the gamma isomer having the highest dipole moment also has the highest insect toxicity, whereas the other isomers have considerably lower dipole moments and accordingly lower toxicity.

In the present study of the series of alkyl esters of *n*-phenyl carbamic acids, it was reasoned that since molar refractivity is a function of the geometric configuration of a molecule in any homologous series, this measurement might offer a clue to the correlation of physical chemical properties with biological activity. Accordingly, the refractive index of this series of compounds was determined with an Abbe refractometer at 91° C \pm 0.1°. The compounds have previously been purified by recrystallization from petroleum ether. The density of the compound was determined by the volume displacement method at 20° C.

The biological activity of the compound was determined by planting the seeds in triplicate in gallon cans that had been previously treated with an amount of material calculated to give 1 lb of active ingredient per acre. Notes were taken on the number of seedlings that emerged, and the plants were harvested after two weeks' growth, and weighed.

From the data derived from the refractive index and density measurements, molar refractivity was calculated according to the Lorentz-Lorenz (12) formula.

$$N = \frac{n^2 - 1}{n^2 + 2} \cdot \frac{M}{d}$$

A further calculation was made by multiplying the molecular refractivity by the factor of the melting point of the derivative divided by the boiling point of the parent alcohol.

$$Q = N \cdot \frac{\text{mp (derivative)}}{\text{bp (alcohol)}}$$

These calculations were made prior to obtaining the data from the toxicity experiment. After obtaining the data

of the toxicity experiments the correlation coefficient was calculated. This was found to be -0.84 which is significant at the 0.01 level, according to Snedecor's table of probability (8), indicating a very high degree of significance and correlation.

Table 1 presents the physical constants of the alkyl derivatives of *n*-phenyl carbamic acid, the boiling point of the parent alcohol, melting point of the derivative, density of the derivative, and refractive index.

Table 2 shows the molecular refractivity, and "Q function" of the phytotoxicity of these alkyl groups, as measured by the growth in milligrams per plant.

TABLE 2

Alkyl group	Molar refractivity at 91° C $\pm 0.1^\circ$ C	Q function	Growth of plants mg/plant
Methyl	40.6	28.9	99.7
Ethyl	53.7	35.8	35.4
Propyl	52.2	29.7	104.2
Isopropyl	50.5	52.2	0.00
Butyl	58.0	29.2	122.3
Sec-butyl	33.2	10.0	124.8
Isobutyl	35.5	26.2	119.3
Amyl	59.9	27.2	121.0
Isoamyl	62.5	26.0	131.0
None (control)	124.3

The formula (8) used for calculating the correlation coefficient is as follows:

$$r_{xy} = \frac{N \sum xy - TxTy}{\sqrt{N(\sum x^2) - (Tx)^2} \sqrt{N(\sum y^2) - (Ty)^2}}$$

It was found that the molecular refractivity itself would not correlate as well with phytotoxicity as did the so-called Q function in this series.

Certain other derivatives were tested, notably those of *m*-chlorophenyl carbamic acid, to see if the Q function would fall within the same range as those by phenyl carbamic acid derivations. This was not found to be the case; therefore, it is reasoned that the values given in the tables apply only to a homologous series. It is interesting to note, however, that certain fundamental physical measurements made to elucidate something of the geometry of the molecule can be correlated with toxicity. If sufficient information appertaining to physical configuration as related to biological activity were amassed, it would enable the biologist and chemist to predict the activity of a compound and prepare biologically potent materials.

In the author's laboratory it has been possible to demonstrate the effect of iso-propyl-*n*-phenyl carbamate on certain enzymes of germinating Gramineae seedlings as evidenced by lack of color or weak color in treated plants compared to untreated plants when using 2,3,5-triphenyl tetrazolium chloride to indicate the reaction. Preliminary evidence also indicates the possibility of reversing the poisoning action of iso-propyl-*n*-phenyl carbamate by meta inositol.

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The Relationship between Human Serum Cholinesterase and Serum Albumin

Milton Gjelhaug Levine and Robert E. Hoyt^{1, 2}

Institute of Experimental Medicine,
College of Medical Evangelists, Los Angeles

In a recent paper (5) we reviewed the literature on the variation of human serum cholinesterase (pseudocholinesterase) in various pathological states and presented our observations on the decrease of this enzyme in patients with malignancy, and in pregnant women. In an attempt to explain this decrease, we have studied further serum cholinesterase levels in numerous diseases and have observed what we consider to be a general principle applicable to this phenomenon.

In the process of explaining the influence of disease on this enzyme, we noticed an apparent correlation between the serum albumin and cholinesterase levels. In order to establish such a relationship we studied the level of these two substances in a variety of diseases, and are presenting here the results obtained with a series of 294 patients seen routinely at our hospital and diagnosed as having the usual variety of diseases to be expected in a group of general hospital admissions.

In tests for serum cholinesterase, we have followed the procedure of Mazur and Bodansky (6), who modified the method of Ammon (1). The production of acetic acid from acetylcholine was followed at 37° C by means of the liberation of carbon dioxide from a bicarbonate-carbonic acid buffer in Warburg flasks. These vessels were gassed at room temperature with a mixture of 93% oxygen and 7% carbon dioxide. At zero time, the acetylcholine was tipped into the serum and bicarbonate mixture. Readings were made at 20 min, at which time the readings fell on a zero order curve. Albumin determinations were made by our own modification of the Biuret method, which we shall describe elsewhere.

In a consecutive series of cases, a large number of dis-

¹ Assisted by Anita A. Suran.

² Aided by a grant from the Cancer Research Grants Division, U. S. Public Health Service.

eases were studied, including pernicious anemia, tuberculosis, leukemia, tumors (benign and malignant), cirrhosis, infectious hepatitis, diabetes, and other conditions. The data to be presented for the series indicates a statistical correlation between cholinesterase and the albumin levels. In each individual case, within the limits of the normal range of variation for both cholinesterase and albumin, it is impossible to establish such a correlation. However, for values below the normal range, it is possible. Thus

CHOLINESTERASE
(μ L CO_2 EVOLVED)

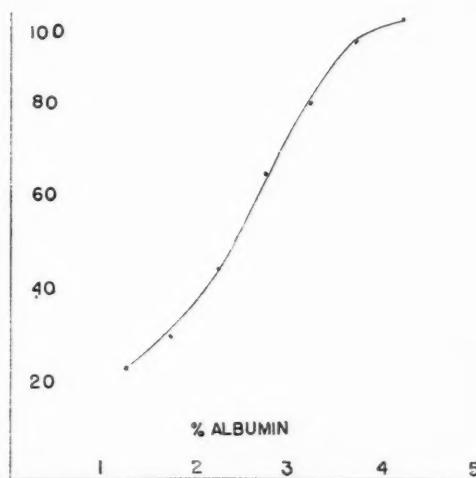


FIG. 1. Cholinesterase and serum albumin.

very low values for cholinesterase were always associated with low albumin values and vice versa. There was only one exception noted. If albumin was low due to increased urinary excretion, the cholinesterase in many cases was normal or higher than normal. In fact, we could predict albuminuria from the presence of a markedly elevated cholinesterase albumin ratio.

The relationship between serum albumin and cholinesterase in patients without albuminuria may be seen in Fig. 1. By dividing patient albumin values into groups varying by 0.5 g of albumin per 100 ml, and then averaging the cholinesterase values for each group, we see the definite relationship between the two. The detailed values for this group are given in Table 1.

Since numerous previous workers have noted the decrease in serum albumin in various diseases including cancer, our finding of the correlation of serum albumin and serum cholinesterase would obviate further work on the level of this enzyme in each individual disease. Rather, research now should be directed toward finding the mechanism involved in the relationship of the serum albumin and cholinesterase.

That the liver is involved in the production of serum cholinesterase may be deduced from the fact that patients with liver damage show a decrease in both albumin and cholinesterase. We have noted this, as have also Kunkel and Ward [4]. Whether the liver damage affects the enzyme directly or indirectly (through its effect on albumin) remains to be determined.

In the same way, decrease in the enzyme in other diseases may be due primarily to decrease in albumin (which decrease may in turn be due to liver damage or protein intake deficiency), or to interference with a mechanism in which albumin hypothetically may help form the en-

TABLE 1
RELATIONSHIP BETWEEN SERUM ALBUMIN AND
SERUM CHOLINESTERASE

Serum albumin g/100 ml	Number of patients	Cholinesterase average value	Cholinesterase standard deviation
1.1-1.5	4	24.0	9.24
1.6-2	13	30.7	8.82
2.1-2.5	23	45.2	18.4
2.6-3	55	65.9	17.8
3.1-3.5	90	80.9	23.4
3.6-4	82	93.6	19.5
4.1-4.5	18	98.7	14.5

zyme, or lastly, to a deficiency in a mechanism which forms both albumin and serum cholinesterase independently of each other.

The fact that normal or high values for serum cholinesterase are associated with low albumin values in some cases of kidney damage (observed also by Kunkel and Ward [4], and by Faber [3]) is to be expected. The smaller molecule of albumin readily passes through the kidney filter, which retains the larger cholinesterase molecule (2). In addition, it has been suggested by a number of workers that in glomerulonephritis there is a compensatory increase in albumin production. The exceptionally high cholinesterase levels encountered in some cases of albuminuria may be explained again on the basis of the correlation noted between serum cholinesterase and albumin production.

It has been demonstrated that the level of serum cholinesterase varies directly with that of serum albumin in various pathological states. There was only one exception noted; a low albumin associated with a normal or high serum cholinesterase is frequently observed in patients with albuminuria.

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Comments and Communications

Foraminifera and Deep Sea Stratigraphy

In a recent paper, Fred B. Phleger, Jr., (*Medd. Öc. Inst. Göteborg*, 1948, P. 16) describes the distribution of pelagic foraminifera in a submarine core, raised by the Swedish Deep Sea Expedition from the Caribbean Sea. The faunal variations are interpreted in terms of relative temperature, and a tentative correlation with the general quaternary stratigraphy of the North American continent is made.

There is, however, another interesting statement in Phleger's paper. He writes:

There were certain samples and groups of samples in this core which contained a very high percentage of broken and somewhat eroded specimens; this occurrence is shown in figure 1. The breakage was not due to the process used in the preparation of the samples for study, since most of the samples contained little or no broken material. The specimens were broken and partially eroded either during or after deposition. The significance of these layers of broken material is not apparent to the writer.

The present author has had the opportunity of studying deep sea cores, and those belonging to the collections of the Swedish Deep Sea Expedition. The phenomenon mentioned has been found to be common to tropical and subtropical deep sea sequences. The zones with broken foraminifera, in all observed cases, were found in a typical petrographic environment. From Phleger's diagram (Plate I), it appears that the "broken specimens" were restricted with two exceptions to zones where deposits were made in warm water. In pelagic sediments, zones rich in crushed shells are characterized by many traces of mud-eating bottom organisms, by a comparatively high concentration of dark organic matter, and by a lowered concentration of calcium carbonate. Some of these relationships were indicated by Roger R. Revelle (*Carnegie Inst. Wash. Publ.* 556, 1944) on a regional basis. Revelle, however, does not try to explain them.

It seems certain that the conditions mentioned indicate epochs when mud-eating organisms extensively reworked the sediment, crushing especially the larger and more fragile shells. The increased influence of the mud-eaters on the sediment may be caused either by a decreased rate of sedimentation or by an increased number of mud-eating organisms at the deep sea bottom. If the ecological conditions at the deep sea bottom change so as to favor a growing number of mud-eaters per surface unit, one is inclined to assume a contemporaneous increase also of other benthonic organisms. The size and structure of the benthonic shells render them fairly resistant against crushing. Their relative abundance is, therefore, strongly increased in the reworked zones. If, however, the amount of benthonic foraminifera is calculated in terms of number per weight unit of sediment, this number seems to be almost constant and identical, both in layers with a low and in those with a high degree of crushing.

The first alternative, therefore, seems to give the most

adequate explanation for the observed conditions. If the rate of sedimentation is low, a layer will be reworked by organisms several times before being covered thickly enough to prevent the penetration of digging animals.

In a pelagic environment the changes in growth of sediment are largely dependent upon variations in the amount of biogenous material, mainly calcareous and siliceous shells, which subside to the bottom. This amount is in turn determined by the production of organisms, especially in the surface layer of the ocean and by the decomposition and dissolution of the subsiding matter. No traces of dissolution have hitherto been observed on entire shells in the zones of crushing. The stratification mentioned is, therefore, thought to be caused mainly by changes in the production of plankton. Periods designated by Phleger as warm are thus characterized by a low production; whereas, during periods which Phleger assumed were cold, the surface layer of the ocean seems to have been more fertile, locally or regionally.

The displacement, during the ice ages, of cold water zones at the divergences of the equatorial current system is thought to be the main cause of changes in distribution of heat in the equatorial surface layer of the ocean.

A closer study of these phenomena is being carried out on sediment cores brought home from the eastern Pacific area by the Swedish Deep Sea Expedition and results will be published in reports of the expedition.

GUSTAF ARRHENIUS

*Kagghamra, Grödinge,
Sweden*

A Folliculinid from Northwestern Iowa

The genus *Folliculina* is one of six genera of sessile, loricate, heterotrich, ciliate Protozoa, placed by A. Kahl in the family *Folliculinidae* Dons (In F. Dahl, ed. *Tierwelt Deutschlands unter der angrenzenden meeressteile*, Jena: Gustav Fischer, 1935). The distribution of the folliculinids has been discussed by E. A. Andrews (*Trans. Amer. micro. Soc.*, 1948, 67, 61). Of about thirty species which have been described for this family, all are marine except *F. boltoni* Kent, which is found in fresh water. Andrews states that *F. boltoni* has been reported from England, France, Switzerland, Uruguay, and questionably from Vancouver Island, British Columbia.

During July and August, 1948, about thirty specimens of a folliculinid were obtained from Little Millers Bay, Lake Okoboji, Dickinson County, Iowa. A few specimens were obtained from the same locality during July 1949. The animals were found on slides which had been suspended in the lake: (1) near the surface less than 1 m from the shore, and (2) near the bottom (depth of about 2 m) about 10 m from the shore. The lake at this point has a sandy bottom with a profuse growth of aquatic plants. The shore is bordered by rocks covered with diatoms, algae, fresh-water sponges, and other sessile and creeping forms.

Although the organism has not been found in sufficient numbers to make a complete study of its life cycle or of its structure, a study of living and of stained specimens leaves no doubt that it is a folliculinid, probably *Folliculina boltoni* Kent. Insofar as can be determined, this is the first unquestionable record of a fresh-water folliculinid from North America, and is the first time that a folliculinid has been found so far from the seacoast.

JOHN MEACHAM HAMILTON

*Park College and the Iowa Lakeside Laboratory,
Parkville, Missouri*

Photographing Graphs for Publication

Gutsell's note (*Science*, 1949, 110, 403) on the preparation of graphs for publication suggests a method which is extremely roundabout and quite unnecessary. He proposes working on the reverse side of graph paper in order to eliminate the graph lines in photographic reproduction of charts.

No high order of photographic skill is required to make use of graph rulings and still eliminate them from photographs. I use Dietzgen millimeter cross-section paper #338, the light green lines of which are held back very well when the desired chart is photographed through a green filter (X2 or X3) onto contrast process film.

The method is simple; if standard lighting is always used, other data are readily standardized and any quantity of charts may be easily and satisfactorily prepared.

HAROLD RAOUL WAINERDI

*Editor, M D
Brooklyn, New York*

Hotchkiss Reaction and Structure of Polysaccharides

A method of staining polysaccharides based on oxidation with periodic acid followed by combination of resulting aldehydes with fuchsin sulfurous acid has been published by Rollin D. Hotchkiss (*Arch. Biochem.*, 1948, 16, 131). The required conditions for a positive reaction are supposed to be two adjacent free hydroxyl groups. The reaction has been used for testing polysaccharides in solution. Jorpes, J. Erik, Werner, Birgitta, and Åberg, Bertil (*J. biol. Chem.*, 1948, 176, 277) used this method in an attempt to detect the presence of such hydroxyl groups in heparin trisulfuric and monosulfuric acids, chondroitin sulfuric acid, and hyaluronic acid.

As far as our experience goes, the presence of two adjacent free hydroxyl groups within the chain of the polysaccharide does not bear any relationship to a positive Hotchkiss reaction. Numerous sugars having such groups, such as cellobiose, methyl α -D-glucopyranoside, methyl n -acetyl- α -D-glucosaminide, give a negative reaction, while hyaluronic acid and chitin, which yield a strong positive reaction, consume only a very small amount of periodic acid (0.1-0.4 mole for each repeating unit). Under the same conditions, starch, glycogen, and cellulose consume one molecule of periodic acid for each pair of adjacent free hydroxyl groups.

Since the information bearing on the mechanism of the Hotchkiss reaction is only fragmentary, we consider it

unsafe to use this reaction for identification of polysaccharide structure.

ROGER JEANLOZ

*The Worcester Foundation for Experimental Biology,
Shrewsbury, Massachusetts*

Categories of Availability or Validity of Zoologic Names

Recent publication by one of us (Smith, H. M., *Science*, 1947, 106, 11) of a note on the use of the expressions *valid* and *available* in describing the status of scientific names prompted the other two to write him that experience in other groups of animals might modify the conclusions that he had reached. Comparison of usage in our three widely separated fields (herpetology, entomology, and paleontology) has led to substantial agreement on a set of terms and definitions different from those previously held by any of us. It is thought that these conclusions may be of interest to others, for the categories involved are not clearly understood by all taxonomists, and the terminology is often confused in practice. Particularly confusing are the uses of *valid* or *validly* by different writers for several of the categories.

Zoological names appear to fall into four categories in respect to their nomenclatural status. (1) All names that have appeared in print (in the broadest sense) must be considered for possible acceptance into scientific nomenclature. (2) Printed names that meet all the publication requirements of the *International Rules of Zoological Nomenclature* are automatically accepted into nomenclature. (3) Names published in full accord with the *Rules* are nomenclaturally acceptable if they are not preoccupied by another name of the same spelling. (4) From among the nomenclaturally acceptable names, there is only one which, because it is the oldest or has been judicially accepted, can be properly used to the exclusion of all others under a given set of circumstances.

The first of these categories generally has not been given a name, although *printed*, *published*, and *occupied* have all been used. We believe that *printed* is not sufficiently descriptive since a printed label should be excluded, and *occupied* implies "in nomenclature" and so is more appropriate in the second category. *Published* appears to be logically applicable to all names that have appeared in print (in the broadest sense). Most published names are accepted into nomenclature, but some fail to meet requirements of the *Rules* and are disregarded in nomenclature; examples are vernacular names, names without referents (*nomina nuda*), and names printed in mediums not qualifying as scientific publications.

Names in the second category have generally been cited as *published*, but *available* has also been used. In the customary sense, however, not all these names are available for use, since some are junior homonyms; and to be exact in this usage, *published* must be modified by "under the *Rules*." The term *occupied* may be applied appropriately to those published names that do meet the requirements of the *Rules* as to publication. *Occupied* names include all names published according to the technical requirements of the *Rules*—all names that are ac-

cepted into zoological nomenclature, such as valid names, synonyms, homonyms, and *nomina inquirendae*.

The word *occupied* in this sense has an unfamiliar ring and may at first glance seem inappropriate. However, if one imagines that there is theoretically a niche for each possible combination of letters that could form a name under the *Rules*, and that when a name is published it would occupy its particular niche, the concept of occupation becomes clearer. Note also that the common use of the familiar term *preoccupied* makes it easier to understand the corresponding term *occupied*.

In the third category names have been described as *valid* or *available*. They are available for use, but they are not valid in the more common sense of that word, as being the one acceptable name. The numerous ways of using *valid* make it unsuitable for a sharply defined concept. We may then apply the term *available* to all names that were published in accordance with the requirements of the *Rules* (legally published) and which have not been so published previously for some other genus of animal or for some other species of the same genus. If any of them has been so published previously, the later name is said to be preoccupied and is called a homonym, or more significantly, a junior homonym, and is not available. Thus all names are available which are now properly in use or which may at any future time be properly used.

Finally the one name under which the species or genus is to be known has also been called the *valid* or *available* name. *Available* seems more appropriate in the third

category, and *valid* has the disadvantage of being commonly used with several meanings. We suggest that the simple and self-expressive term *correct* be applied in a technical sense to the oldest available name for a genus or for a species within a genus. If the oldest available name has been set aside by the International Commission using its plenary powers, then the next oldest name or the one designated by the commission is the *correct* name.

The categories may then be defined and named as follows, according to these conclusions: Any name that is printed and circulated is *published*; any published name that meets the publication requirements of the *Rules* is *occupied* in zoological nomenclature (if it fails to meet the requirements it is an outlaw name, i.e., unpublished, illegally published, or a *nomen nudum*); any occupied name that is not preoccupied by an older name of the same spelling is *available* (if it is preoccupied it is a junior homonym and is not available); the oldest available name is the *correct* name, unless it has been specifically set aside by the commission under the plenary powers. (The *correct* name will, of course, vary with changes in our knowledge of subjective synonymy or discovery of unknown facts in the history of the names, such as homonymy and objective synonymy. An available name whose genus cannot be identified is a *nomen dubium*.)

RICHARD E. BLACKWELDER, J. BROOKES KNIGHT,
and HOBART M. SMITH

U. S. National Museum,
Washington, D. C.

Scientific Book Register

The Meaning of Relativity. 3rd ed. including "The Generalized Theory of Gravitation." Albert Einstein. Princeton, N. J.: Princeton Univ. Press, 1950. 150 pp. \$2.50.

Microbiologie du Sol: Problèmes et Méthodes. S. Winogradsky. Paris VI^e: Masson et Cie, 1949. 861 pp.; illustrated. 3000 fr.

Plant Pathology. Sir Edwin J. Butler and S. G. Jones. London-New York: Macmillan, 1949. 979 pp.; illustrated. \$10.00.

Biophysical Research Methods. Fred M. Uber, Ed. New York-London: Interscience, 1950. 667 pp.; illustrated. \$9.50.

Adaptation and Origin in the Plant World: The Role of Environment in Evolution. Frederic E. Clements, Emmett V. Martin and Frances L. Long. Waltham, Mass.: Chronica Botanica; New York: Stechert-Hafner, 1950. 332 pp.; illustrated. \$6.00.

Biological Actions of Sex Hormones. Rev. 2nd ed. Harold Burrows. New York 10: Cambridge Univ. Press, 1949. 615 pp. \$8.50.

Nomenclator Zoologicus, Vol. V, 1936-1945. Sheffield A. Nave, Ed. London N.W.8: Zoological Society of London, 1950. 308 pp. £3 13s. 6d.

Knowing and the Known. John Dewey and Arthur F. Bentley. Boston: Beacon Press, 1949. 334 pp. \$4.00.

Science in Education. M. C. Nokes. London-New York 10: Maedonald & Co., 1949. 158 pp. \$2.00.

Electromagnetic Theory. Proceedings of Symposia in Applied Mathematics, Vol. II. New York 27: American Mathematical Society, 1950. 91 pp.; illustrated. \$3.00.

Foundry Science: Fundamentals underlying Foundry Practice. Harry A. Schwartz. New York 19: Pitman Publ., 1950. 286 pp.; illustrated. \$6.50.

Measure Theory. Paul R. Halmos. New York: D. Van Nostrand, 1950. 304 pp. \$5.90.

Frontiers in Colloid Chemistry, Vol. VIII. R. E. Burk and Oliver Grunmitt. New York: Interscience, 1950. 157 pp.; illustrated. \$4.00.

Plane and Spherical Trigonometry. M. Richardson. New York: Macmillan, 1950. 343 pp. + 138 pp. logarithmic and trigonometric tables; illustrated. \$3.75.

Comparative Anatomy Laboratory Manual. Lloyd Raymond Gribble. Philadelphia-Toronto: Blakiston, 1950. 231 pp.; illustrated. \$3.00.

Structure and Development of the Vertebrates: A Manual for an Integrated Course in Comparative Anatomy and Embryology. Florence Moog. New York: Prentice-Hall, 1949. 170 pp.; illustrated.

The Physical World. Paul McCorkle. Philadelphia: Blakiston, 1950. 450 pp.; illustrated. \$4.25.

Algebraic Curves. Robert J. Walker. Princeton, N. J.: Princeton Univ. Press, 1950. 201 pp.; illustrated. \$4.00.

NEWS and Notes

Otto Struve has been appointed professor in astrophysics and chairman of the Department of Astronomy of the University of California at Berkeley, effective July 1. He will replace **Sturla Einarsson**, retiring chairman. Dr. Struve, who is best known for his work on interstellar matter and the spectra of stars, was chairman of the Department of Astronomy and director of Yerkes Observatory, University of Chicago, and also director of the McDonald Observatory, operated by the Universities of Chicago and Texas.

Raymond N. Keller, of the University of Michigan, has joined the staff of the Argonne National Laboratory as senior chemist. He will work on cooperative research with Michigan.

John P. Hubbard has been appointed George S. Pepper Professor of Public Health and Preventive Medicine and chairman of the department at the University of Pennsylvania School of Medicine. Dr. Hubbard was formerly director of the Study of Child Health Services, American Academy of Pediatrics.

Edward B. Tuohy, professor of anesthesiology, Georgetown University Medical School, Washington, D. C., will present a paper on "The Management of Hypertensive Episodes During Anesthesia, Including a Discussion of the Entity Pheochromocytoma" at the Southern Association of Anesthesiologists meeting in St. Louis, March 31-April 1.

Raymond Gautier, assistant director general of the World Health Organization, retired January 31, to accept the post of research director of the new International Children's Center in Paris. Dr. Gautier was associated with the League of Nations health organization for 20 years and later served as director of the Geneva office of the WHO Interim Commission.

Arthur Adel, professor of physics at Arizona State College, Flagstaff, will be one of the speakers at the centenary meeting of the Royal Meteorological Society at Oxford and London, March 28-April 3.

Douglas H. K. Lee, professor of physiological climatology in the Isaiah Bowman School of Geography, and lecturer in physiological hygiene in the School of Public Health and Hygiene, Johns Hopkins University, will deliver the Cutter Lecture on preventive medicine at Harvard Medical School on April 24. His subject will be "Physiology as a Guide to Combating Tropical Stress."

Visitors to U. S.

B. N. Narayana Iyengar, lecturer in electrical engineering, Indian Institute of Science, Bangalore; **Joel Lindberg**, director of applied research, Swedish Institute of Textile Research, Gothenburg; and **D. Tollenaar**, chief of the Physicochemical Department, Research Institute for the Graphic and Allied Industries, T.N.O., Amsterdam, were recent visitors at the National Bureau of Standards.

Industrial Laboratories

The A. H. Robins Company, Inc., Richmond, Virginia, has appointed **Robert S. Murphy**, organic chemist, to its research staff, where he will be in charge of organic synthesis. Dr. Murphy was formerly with the National Cancer Institute.

The W. M. Welch Manufacturing Company, 1515 Sedgwick Street, Chicago 10, is distributing on request copies of a chart of the radioactive elements, prepared by **Herta R. Leng** of Rensselaer Polytechnic Institute.

Grants and Awards

The National Cancer Institute announced on March 6 the award of Public Health Service grants of \$458,476 to 20 medical schools and \$63,705 to 13 dental schools to support cancer teaching. Among the medical school awards one new grant of \$25,000 was made to Indiana Uni-

versity Medical Center, Indianapolis. The others were renewal grants. Twelve renewal grants were made to dental schools and one new grant of \$5,000 to the University of Washington School of Dentistry, Seattle.

Grants totaling \$575,100 for construction of cancer research facilities were also announced, including five new grants and two supplemental grants. The new grants are: North Carolina University School of Medicine, Chapel Hill—\$200,000; Medical College of the State of South Carolina, Charleston—\$100,000; The Children's Medical Center, Boston—\$100,000; Beth Israel Hospital, Boston—\$50,000; State University of Iowa College of Medicine, Radiation Research Laboratory, Iowa City—\$12,250. Supplemental grants were made to Wayne University College of Medicine and Detroit Institute of Cancer Research, and the Boston University School of Medicine.

The John and Mary R. Markle Foundation has named 20 medical scientists as the third group of Scholars in Medical Science (see *Science* August 12, 1949, p. 173). Five hundred thousand dollars for their support will be allotted in grants of \$25,000 each, at the rate of \$5,000 a year to the medical schools with which they are associated. Under the program, now in its third year, a total of 48 doctors will be aided with grants totaling \$1,200,000. The grants are made "for support of outstanding young scientists who have chosen academic medicine in preference to practice or research in industrial laboratories. The purpose is to afford them academic and financial security to develop at their own pace."

The Scholars, their fields of interest, and the schools receiving grants for their support are: **Georges-Albert Bergeron**, pathological physiology, Laval University Faculty of Medicine, Quebec; **Charles G. Craddock, Jr.**, emotional aspects of disease, University of Virginia Department of Medicine; **Thomas Timothy Crocker**, infectious diseases, University of California Medical School, San Francisco; **Quentin B. Deming**, heart and kidney disease, Stanford University School

of Medicine; *Francisco Garcia-Benocera*, neurosurgery and neurology, Tulane University of Louisiana School of Medicine; *Robert Alan Good*, pediatrics (rheumatic fever), University of Minnesota Medical School; *Robert L. Grissom*, cardiovascular disease, University of Illinois College of Medicine; *Daniel Lewis Larson*, immunochemistry in cancer, Columbia University College of Physicians and Surgeons; *John Phillip McGovern*, pediatrics (neonatal period), George Washington University School of Medicine; *Robert James McKay, Jr.*, pediatrics (heart disease, physiologic anemia of infancy), University of Vermont College of Medicine; *Gardner C. McMillan*, degenerative diseases, McGill University Faculty of Medicine; *Samuel Preston Martin*, infectious diseases, Duke University School of Medicine; *Lawrence Peters*, pharmacology and therapeutics, Western Reserve University School of Medicine; *Frank William Putnam*, physical chemical properties of proteins and viruses, University of Chicago Division of Biological Sciences; *William B. Schwartz*, cardiovascular disease, Tufts College Medical School; *Joseph E. Sokal*, internal medicine and oncology, Yale University School of Medicine; *Lloyd Grenfell Stevenson*, medical history and preventive medicine (history of disease), University of Western Ontario Faculty of Medicine; *Wade Volwiler*, liver disease, University of Washington School of Medicine, Seattle; *James Walker, Jr.*, surgery (therapy of burns and shock), University of Pennsylvania School of Medicine; and *Ernest Harshaw Yount, Jr.*, degenerative and metabolic disease, Bowman Gray School of Medicine, Wake Forest College.

The appointments begin this year.

Edward G. Hampp, of the Oral Bacteriological Section, National Institute of Dental Research, received the 1949 Washington Academy of Sciences award in the biological sciences on March 16. Dr. Hampp was honored for his contributions to the knowledge of diseases of the oral cavity, and particularly because he was the first to isolate *Borrelia vincentii* in pure culture.

Fellowships and Prizes

The Research Corporation of New York has made a grant of \$5,500 to Polytechnic Institute of Brooklyn in support of a \$6,800 research project (1950-51) on the study of plate efficiency in distilling columns, under the direction of Ju Chin Chu, associate professor of chemical engineering. Two \$2,000 predoctoral fellowships for the project are now available. Qualified chemical engineers are invited to submit their applications to Prof. Donald F. Othmer, head of the Chemical Engineering Department, Polytechnic Institute of Brooklyn, before April 10.

The American College of Chest Physicians is offering an annual international award of \$250 for the best original paper on any chest disease. Five copies of the manuscript should be submitted not later than May 1 to the Executive Secretary, American College of Chest Physicians, 500 North Dearborn Street, Chicago 10. Further information may be obtained from the secretary.

An international contest for an original and unpublished article on the physiopathology, clinical history, and therapy of rheumatic and arthritic diseases is being sponsored by the Azienda Autonoma of the Health Resort of Acqui, Italy. A prize of one million lire will be awarded. Papers must be submitted by December 31. Further information can be obtained from the Azienda Autonoma Di Cura Acqui, Piemonte, Italy.

Colleges and Universities

Yale University is conducting a series of special lectures in applied statistics February 7-May 18. Each visiting lecturer will be available for a limited number of individual or group conferences, and the lectures and Thursday evening seminars are open to all interested. Further details may be obtained from Mrs. S. H. Donahue, Room 1247A, Trumbull College, or from C. I. Bliss or D. F. Votaw, Jr., Department of Mathematics, Yale University, New Haven, Connecticut.

The University of Oklahoma Seminar on Social Psychology at Crossroads 1950 will be in session April 6-11, at the Extension Study Center, Norman, Oklahoma. Leading research men in the fields of biology, psychology, anthropology, sociology, and other social sciences will meet to discuss methodological problems of psychological organization at various phylogenetic levels, of cross-cultural comparisons, the implications of recent developments on judgment, perception, learning, ego functioning, language development, group relations, and personality.

The University of Pennsylvania Museum is sponsoring an archaeological expedition to the former city of Gordian, in Turkey, to search for objects and material which will provide more information on Phrygian civilization and culture from the tenth to the seventh centuries B.C. Rodney S. Young, curator of the Mediterranean Section of the museum, who will head the expedition, is now in Turkey making preliminary arrangements. Digging is scheduled to begin early next month and continue until mid-July. It is hoped that material uncovered will help determine the extent of Phrygian influence on later Greek civilization.

Meetings and Elections

The Metropolitan Section of the American Physical Society will hold its spring meeting March 31-April 1 at the Brookhaven National Laboratory. There will be a technical program of invited papers on both days. Admission to the entire meeting or to a single session may be obtained only by sending advance notice to Marriette Kuper, Brookhaven National Laboratory, Upton, Long Island, New York.

The annual Colloquium of College Physicists will be held June 14-17 at the State University of Iowa, Iowa City. The program will include research lectures by H. C. Urey, Katherine B. Blodgett, John Spence, and Robert L. Sinsheimer. Round table discussions of laboratory and of physics history for non-science students will be led by Duane Roller. Four lectures on hydrodynamics in cosmic physics are to

be presented by Edward Teller, under the sponsorship of the Research Foundation. Requests for final programs should be sent to G. W. Stewart at the university.

The Sixth International Congress on the History of Science will be held in Amsterdam, August 14-21. A special section on the history of medicine will constitute the **12th Congress of the Société Internationale d'Histoire de la Médecine**. Papers to be presented at the congress must be submitted before **May 1**. Full information may be obtained from Prof. Ir. R. J. Forbes, Haringvlietstraat 1, Amsterdam-Z, The Netherlands.

The Committee on Human Reproduction of the National Research Council will hold its annual two-day conference on reproduction at the Hotel Commodore, New York City, May 22-23. The subject this year will be testis and ovary, eggs and sperm. Among the speakers will be David W. Bishop, University of Massachusetts; L. C. Dunn, Herbert Elftman, and Earl T. Engle, Columbia University; Helen Deane, Harvard Medical School; Henry A. Lardy, University of Wisconsin; John McLeod, Cornell Medical College; Warren O. Nelson, University of Iowa; Jack Schultz, Cancer Research Institute, Philadelphia, and George D. Snell, Jackson Memorial Laboratory, Bar Harbor, Maine.

A conference on ionospheric physics, sponsored by Pennsylvania State College and the Geophysical Research Directorate of the U. S. Air Force, will be held at the college July 24-26. Papers will be presented on the latest theoretical and experimental developments in the field of physics relative to the upper atmosphere. Speakers from several foreign countries are expected to participate. Further details may be obtained from A. H. Waynick, Radio Propagation Laboratory, Pennsylvania State College, State College, Pennsylvania.

The American Academy of Orthopaedic Surgeons elected the following officers on February 16: president elect 1951-52, Joseph H. Barr, Boston; vice president, J.

Hiram Kite, Atlanta; secretary, Harold B. Boyd, Memphis; treasurer, H. Relton McCarroll, St. Louis; and librarian-historian, Claude N. Lambert, Chicago.

Officers elected at the annual meeting of the board of trustees of **Biological Abstracts**, in Philadelphia, February 19, were: president, Maurice B. Visscher, University of Minnesota; vice president, Stuart Mudd, University of Pennsylvania; treasurer, J. J. Willaman, Eastern Regional Research Laboratory; and secretary, Robert Gaunt, Syracuse University.

Deaths

Charles J. Pierson, 83, head of the Department of Zoology of Maryland University from 1921 until his retirement in 1937, died February 28, at Laurel, Maryland, after a long illness. Dr. Pierson's contributions to zoology included collection of rare fish specimens in the Pacific, a biological survey of the Bay of Panama, and the discovery of fossils in the plains of Kansas.

Percy Rogers Howe, director of the Forsyth Dental Infirmary for Children since 1917, died at his home in Belmont, Massachusetts, February 28, at the age of 85. Dr. Howe was credited with perfecting the ammoniacal silver nitrate solution for treatment of dental caries with S. Bert Wolbach, and made extensive studies of the effect of diet on the teeth.

Alfred Habdank Korzybski, 70, originator of the methodology of general semantics, died March 1, in Sharon, Connecticut, of coronary thrombosis. His work, concerned with scientific analysis of blocks to communication and understanding, has been found useful in clarifying many scientific problems, including some issues in mathematical theory.

Horace W. Gillett, 66, international authority on metallurgy, died in Nicholasville, Kentucky, March 3, while returning from a hunting trip in Florida and Georgia. Dr. Gillett was responsible for Battelle Memorial Institute's technical organization, and he served as its first director, from 1929 to 1934. He re-

tired in 1949 as chief technical advisor of the institute.

Miscellaneous

The Viral and Rickettsial Registry, a catalogue of prepared specimens of selected viruses and rickettsiae ready for distribution, is available upon request from the American Type Culture Collection, 2029 M Street, N.W., Washington 6, D. C. The catalogue includes the histories of the specimens, costs, methods of shipment, and other pertinent data.

The Veterans Administration, after three years of controlled testing and development, is making available a **suction socket artificial leg** to eligible veterans whose above-knee amputations are of such character to make use of the device medically feasible. During the past two years, the administration has tested more than 500 amputees in its experimental program. Successful results were obtained in a majority of the cases. The suction socket limb has no pelvic hinge or suspension harness, and it provides greater freedom of movement and a more life-like appearance.

The Veterans Administration's research to determine possibilities and limitations of this limb was done with the joint cooperation of the Advisory Committee on Artificial Limbs of the National Research Council, the University of California, the Orthopedic Appliance and Limb Manufacturers Association, the Army, the Navy and other interested organizations.

Amputee veterans who are eligible for a new artificial leg may obtain complete information from the Prosthetic and Sensory Aids Unit of the nearest Veterans Administration regional office.

The estate of the late H. W. Norris, of Grinnell College, Iowa, offers copies of the paper on **The Plagiotostome Hypophysis**, bound in cloth, to any interested person. Requests should be addressed to Henry S. Conrad, Grinnell College, Grinnell, Iowa.

Pi Mu Epsilon, honorary mathematical fraternity, is now publishing the *Pi Mu Epsilon Journal*, which

will be of interest to both undergraduate and graduate members of the fraternity. Information can be obtained from Ruth W. Stokes, Editor, 15 Smith Hall, Syracuse University, Syracuse 10, New York.

A proposal for an international society of plant taxonomists was presented to the New York meeting of the Systematic Section of the Botanical Society of America and the American Society of Plant Taxonomists by F. R. Fosberg, Catholic University botanist, on December 29.

Dr. Fosberg reported that at present there is no continuing organization of taxonomists equipped to handle such matters as rules of nomenclature, indexing and abstracting services, photographing of type specimens, microfilming of literature, and exchange between countries of specimens, literature, and scientists. In order to carry on such activities there is an urgent need for a strong, international, cooperative organization with a paid secretariat of one or more active taxonomists.

The idea for such an organization is not new. At the 1930 Botanical Congress held in Cambridge, the late H. M. Hall, of the University of California, suggested it, and in 1935 at the Amsterdam Congress, E. D. Merrill, professor emeritus of botany at Harvard University, made a similar suggestion. Other proposals have more recently been made by Frans Verdoorn, secretary of the Botanical Section of the International Union of Biological Sciences, and J. Lanjouw, director of the Botanical Museum and Herbarium of the University of Utrecht. Professor Lanjouw's proposal, which is being published in *Chronica Botanica*, Vol. 12, No. 1, will be considered at the Stockholm Congress this summer and Dr. Fosberg hopes that U. S. institutions to be represented at the congress will instruct their delegates to support the formation of such an organization and that they will become active members if the society is formed.

The Canadian Mathematical Congress recently published a pamphlet *Why Study Mathematics?* for

the special use of secondary school teachers and students concerned with vocational guidance. Copies may be obtained from congress headquarters in the Engineering Building, McGill University, Montreal, Canada.

The first issue of the quarterly, *Journal of the Philadelphia General Hospital*, has been published by the hospital. It is edited by P. F. Lucehesi, superintendent and medical director, and a consulting board of editors. Copies of the magazine may be ordered from S. O. Waife, associate editor, Philadelphia General Hospital, 34th and Curie Avenue, Philadelphia 4.

About 250 agricultural leaders from 28 foreign countries will come to the U. S. this year for observation, study, and training. Agencies of the U. S. Department of Agriculture, in cooperation with the Department of State, the U. S. Army in Japan and Austria, and the Economic Cooperation Administration, are developing plans for foreign trainees to study and work in fields of special interest to them. Arrangements have been made for training in laboratories, field stations, and at state colleges and experiment stations, as a means of sharing technical agricultural knowledge. Periods of training will range from 60 days to a year.

The Influenza Information Center at the National Institutes of Health, Bethesda, Maryland, will be continued another year. The center was set up a year ago to serve as Western Hemisphere headquarters for a world-wide program to study influenza and its control. The program was initiated by the World Health Organization and is sponsored in this country by the surgeons general of the Army, Navy, Air Force, and Public Health Service. More than 50 laboratories throughout the country that collaborated in the program last year have been invited to participate again and other laboratories, equipped for influenza work during the past year, have been asked to join the project.

The Registry of Rare Chemicals, 35 West 33rd Street, Chicago 16,

Illinois, lists the following wanted chemicals: vanadium nitride, uranium tetrachloride, zirconium bisulfate, gallium dichloride, carbon monosulfide, cyclohexanethiol, 1-thiahexylbenzene, 1-thiaethylcyclohexane, thiacyclooctane, 2-thianno-10-nadecanone, benzoyltri-fluoroacetone, laurone, prodigiosin, L-atechin, anthrarin, ergothioneine, azoisobutyrodinitrile, canadine, and D-laurodanosine.

Sixty-nine declassified and unclassified reports from Atomic Energy Laboratories, abstracted by the Atomic Energy Commission in January, are now available. These include 16 on biology and medicine; 23 on chemistry; 1 on engineering; 5 on mineralogy, metallurgy, and ceramics; and 24 on physics. A complete list of the reports and information on how to obtain them may be had by writing the Documents Sales Agency, Atomic Energy Commission, Box 62, Oak Ridge, Tennessee.

Make Plans for—

American Society of Pediatrics, eastern area meeting, March 30, Bellevue-Stratford Hotel, Philadelphia.

National Speleological Society, 7th annual convention, March 31-April 2, Dodge Hotel, Washington, D. C.

National Association of Corrosion Engineers, April 4-7, St. Louis, Missouri.

American Association of Anatomists, annual meeting, April 5-7, New Orleans, Louisiana.

Association of American Geographers, annual meeting, April 5-8, Clark University, Worcester, Massachusetts.

Seismological Society of America, annual meeting, April 7-8, Seattle, Washington.

American Society of Mechanical Engineers, spring meeting, April 12-15, Hotel Statler, Washington, D. C.

American Chemical Society, 2nd session of 177th national meeting, April 9-13, Philadelphia; 3rd session, Detroit, April 16-20.



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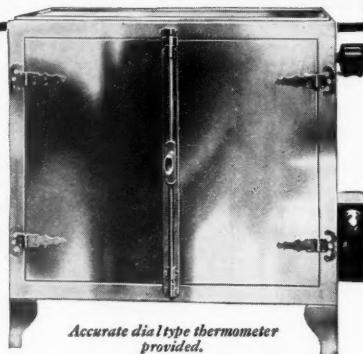
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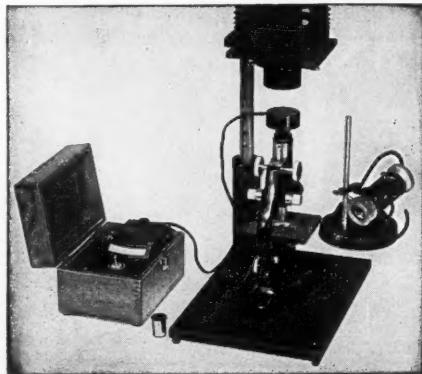
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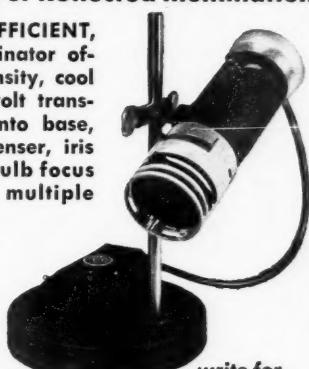
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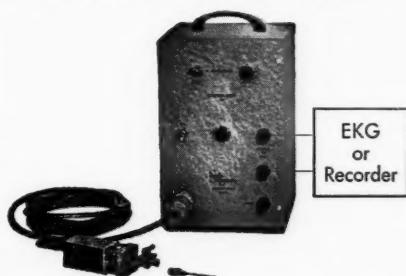
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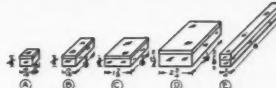
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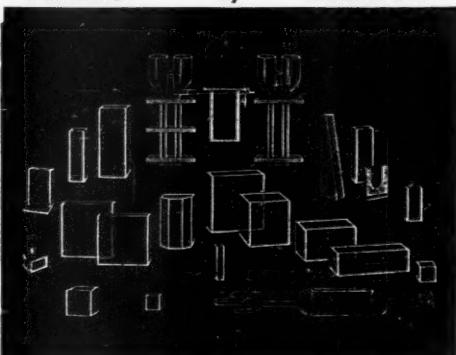
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